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# PROCEEDINGS

## OF THE

# NATIONAL ACADEMY OF SCIENCES

## INDIA

Part 1

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Volume 7

### THEOREMS CONNECTING DIFFERENT CLASSES OF SELF-RECIPROCAL FUNCTIONS

BY B. M. MEHROTRA AND R. V. SHASTRY

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1. Profs. Hardy and Titchmarsh in their joint paper entitled "Formulae connecting different classes of self-reciprocal functions"<sup>1</sup> gave some rules to construct self-reciprocal functions for transforms of different orders from a given function which was reciprocal for a transform of a particular order. Some of their results have been generalised and some new theorems of a similar nature have now been added by Bailey<sup>1</sup> and Mehrotra.<sup>5,7</sup> In this note we give a few more theorems of the same type. The interest of this note lies mainly in the results and not in a rigorous proof thereof. Hence no attempt is herein made to give a rigorous proof and only the formal procedure is given.

Following Hardy and Titchmarsh we shall say that a function is  $R_v$  if it is self-reciprocal for  $J_v$  transforms.

We will make use of the following result due to Hardy and Titchmarsh.<sup>8</sup>

A necessary and sufficient condition that a function  $f(x)$  should be  $R_\mu$  is that it should be of the form

$$f(x) = \frac{1}{2\pi i} \int_{c-i\infty}^{c+i\infty} 2^{\frac{1}{2}s} \Gamma\left(\frac{1}{4} + \frac{1}{2}\mu + \frac{1}{2}s\right) \psi(s) x^{-s} ds, \dots \dots \dots (1)$$

where  $0 < c < 1$

$$\text{and} \quad \psi(s) = \psi(1-s) \quad \dots \quad \dots \quad \dots \quad (2)$$

2. Theorem. If  $f(x)$  is  $R_\mu$  and  $K(x)$  is  $R_\nu$  the function

$$g(x) = \int_0^\infty P(xy) dy \int_0^\infty K(z) f(yz) dz$$

is  $R_\theta$  provided that

$$P(x) = \frac{1}{2\pi i} \int_{k-i\infty}^{k+i\infty} 2^{\frac{1}{2}s} \left\{ \Gamma\left(\frac{1}{4} + \frac{1}{2}\mu + \frac{1}{2}s\right) \Gamma\left(\frac{3}{4} + \frac{1}{2}\nu - \frac{1}{2}s\right) \right. \\ \left. \times \Gamma\left(\frac{1}{4} + \frac{1}{2}\theta + \frac{1}{2}s\right) \chi(s) \right\} x^{-s} ds, \quad \dots \quad (3)$$

where  $\kappa$  is a positive proper fraction and  $\chi(s)$  satisfies (2).

$$\text{Let } Q(y) = \int_0^\infty K(z) f(yz) dz.$$

It has been shown by Mehrotra<sup>6</sup> that

$$Q(y) = \frac{1}{2\pi i} \int_{1-c-i\infty}^{1-c+i\infty} \Gamma\left(\frac{1}{4} + \frac{1}{2}\mu + \frac{1}{2}s\right) \Gamma\left(\frac{3}{4} + \frac{1}{2}\nu - \frac{1}{2}s\right) w(s) y^{-s} ds, \quad \dots \quad (4)$$

where  $w(s) = w(1-s)$ .

Hence by (4) we have

$$g(x) = \frac{1}{2\pi i} \int_0^\infty P(xy) dy \int_{1-c-i\infty}^{1-c+i\infty} \Gamma\left(\frac{1}{4} + \frac{1}{2}\mu + \frac{1}{2}s\right) \times \Gamma\left(\frac{3}{4} + \frac{1}{2}\nu - \frac{1}{2}s\right) w(s) y^{-s} ds, \\ = \frac{1}{2\pi i} \int_{1-c-i\infty}^{1-c+i\infty} \Gamma\left(\frac{1}{4} + \frac{1}{2}\mu + \frac{1}{2}s\right) \Gamma\left(\frac{3}{4} + \frac{1}{2}\nu - \frac{1}{2}s\right) w(s) ds. \quad \int_0^\infty P(xy) y^{-s} dy$$

provided that the inversion of the order of integration is justified.

Therefore

$$g(x) = \frac{1}{2\pi i} \int_{1-c-i\infty}^{1-c+i\infty} \Gamma\left(\frac{1}{4} + \frac{1}{2}\mu + \frac{1}{2}s\right) \Gamma\left(\frac{3}{4} + \frac{1}{2}\nu - \frac{1}{2}s\right) w(s) x^{s-1} ds \\ \times \int_0^\infty P(w) w^{-s} dw.$$

Now, by Mellin's Inversion formulae,<sup>2</sup> from (3) we have

$$\int_0^{\infty} P(w) w^{s-1} dw = 2^{\frac{1}{2}s} \Gamma\left(\frac{1}{4} + \frac{1}{2}\theta + \frac{1}{2}s\right) \Gamma\left(\frac{1}{4} + \frac{1}{2}\mu + \frac{1}{2}s\right) \\ \times \Gamma\left(\frac{3}{4} + \frac{1}{2}\nu - \frac{1}{2}s\right) \chi(s)$$

Changing  $s$  into  $1-s$  we get

$$\int_0^{\infty} P(w) w^{-s} dw = 2^{\frac{1}{2}-\frac{1}{2}s} \Gamma\left(\frac{3}{4} + \frac{1}{2}\theta - \frac{1}{2}s\right) \Gamma\left(\frac{3}{4} + \frac{1}{2}\mu - \frac{1}{2}s\right) \\ \times \Gamma\left(\frac{1}{4} + \frac{1}{2}\nu + \frac{1}{2}s\right) \chi(s).$$

Hence

$$g(x) = \frac{1}{2\pi i} \int_{1-c-i\infty}^{1-c+i\infty} 2^{\frac{1}{2}-\frac{1}{2}s} \Gamma\left(\frac{3}{4} + \frac{1}{2}\theta - \frac{1}{2}s\right) \Gamma\left(\frac{3}{4} + \frac{1}{2}\mu - \frac{1}{2}s\right) \\ \times \Gamma\left(\frac{1}{4} + \frac{1}{2}\nu + \frac{1}{2}s\right) \Gamma\left(\frac{1}{4} + \frac{1}{2}\nu + \frac{1}{2}s\right) \\ \times \Gamma\left(\frac{3}{4} + \frac{1}{2}\nu - \frac{1}{2}s\right) \chi(s) w(s) x^{s-1} ds.$$

Changing  $s$  into  $1-s$  we get

$$g(x) = \frac{1}{2\pi i} \int_{c-i\infty}^{c+i\infty} 2^{\frac{1}{2}s} \Gamma\left(\frac{1}{4} + \frac{1}{2}\theta + \frac{1}{2}s\right) \Gamma\left(\frac{1}{4} + \frac{1}{2}\mu + \frac{1}{2}s\right) \\ \times \Gamma\left(\frac{3}{4} + \frac{1}{2}\nu - \frac{1}{2}s\right) \Gamma\left(\frac{3}{4} + \frac{1}{2}\mu - \frac{1}{2}s\right) \\ \times \Gamma\left(\frac{1}{4} + \frac{1}{2}\nu + \frac{1}{2}s\right) w(s) \chi(s) x^{-s} ds \\ = \frac{1}{2\pi i} \int_{c-i\infty}^{c+i\infty} 2^{\frac{1}{2}s} \Gamma\left(\frac{1}{4} + \frac{1}{2}\theta + \frac{1}{2}s\right) \psi(s) x^{-s} ds.$$

$$\text{where } \psi(s) = \Gamma\left(\frac{1}{4} + \frac{1}{2}\mu + \frac{1}{2}s\right) \Gamma\left(\frac{3}{4} + \frac{1}{2}\omega - \frac{1}{2}s\right) \\ \times \Gamma\left(\frac{1}{4} + \frac{1}{2}\nu + \frac{1}{2}s\right) \Gamma\left(\frac{3}{4} + \frac{1}{2}\nu - \frac{1}{2}s\right) \chi(s) \omega(s)$$

As  $\psi(s)$  satisfies the equation

$$\psi(s) = \psi(1-s)$$

it follows from (1) that  $g(x)$  is  $R_{\theta}$ ,



3. If we take  $\mu = v$  we have

$$\begin{aligned} P(x) &= \frac{1}{2\pi i} \int_{\kappa - i\infty}^{\kappa + i\infty} 2^{\frac{1}{2}s} \Gamma\left(\frac{1}{4} + \frac{1}{2}\mu + \frac{1}{2}s\right) \Gamma\left(\frac{3}{4} + \frac{1}{2}\mu - \frac{1}{2}s\right) \\ &\quad \times \Gamma\left(\frac{1}{4} + \frac{1}{2}\theta + \frac{1}{2}s\right) \chi(s) x^{-s} ds \\ &= \frac{1}{2\pi i} \int_{\kappa - i\infty}^{\kappa + i\infty} 2^{\frac{1}{2}s} \left(\frac{1}{4} + \frac{1}{2}\theta + \frac{1}{2}s\right) \chi'(s) x^{-s} ds \end{aligned}$$

where  $\chi'(s) = \Gamma\left(\frac{1}{4} + \frac{1}{2}\mu + \frac{1}{2}s\right) \Gamma\left(\frac{3}{4} + \frac{1}{2}\mu - \frac{1}{2}s\right) \chi(s)$

As  $\chi'(s)$  satisfies the equation

$$\chi'(s) = \chi'(1-s)$$

it follows that  $P(x)$  is also  $R_\theta$ . Also it has been shown by Mehrotra<sup>6</sup> that if  $K(x)$  and  $f(x)$  are both  $R_\nu$  then

$$\int_0^\infty K(z) f(yz) dz = \int_0^\infty f(z) K(yz) dz.$$

Hence we arrive at the following corollary:—

If  $K(x)$  and  $f(x)$  are both  $R_\nu$  and

$P(x)$  is  $R_\theta$ , the function

$$\begin{aligned} g(x) &= \int_0^\infty P(xy) dy \int_0^\infty k(z) f(yz) dz \\ &= \int_0^\infty P(xy) dy \int_0^\infty f(z) k(yz) dz \end{aligned}$$

is also  $R_\theta$ .

Example:—Let  $K(x) = f(x) = z^{\frac{1}{2} + \nu} e^{-\frac{1}{2}z^2}$ .

$$\begin{aligned} \int_0^\infty K(z) f(yz) dz &= \int_0^\infty z^{\frac{1}{2} + \nu} e^{-\frac{1}{2}z^2 (y^2 + 1)} (yz)^{\nu + \frac{1}{2}} dz \\ &= 2^\nu \Gamma(\nu + 1) \frac{y^{\frac{1}{2} + \nu}}{(y^2 + 1)^{\nu + 1}} \dots \end{aligned}$$

If, now  $P(x) = \frac{1}{x^{\frac{1}{2}}}$  which is  $R_\mu$  we have

$$\begin{aligned} g(x) &= 2^\nu \Gamma(\nu + 1) \int_0^\infty \frac{(xy)^{-\frac{1}{2}} y^{\frac{1}{2} + \nu}}{(y^2 + 1)^{\nu + 1}} dy \\ &= 2^{\nu - 1} \left\{ \Gamma\left(\frac{1}{2}\nu + \frac{1}{2}\right) \right\}^2 x^{-\frac{1}{2}} \end{aligned}$$

which shows that  $g(x)$  is  $R_\mu$ .

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# ON THE DIRECT FORMATION OF IODIDES AND THE DISTANCE OF THE CLOSEST APPROACH OF ATOMS OF IODINE

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## SUMMARY

It has been put forward in the present paper that for any action there is a particular limiting value for the distance of the closest approach of atoms such that if an element has, for the distance of the closest approach of its atoms, a value lower than this limiting value, it would take part in the action. This limiting value has been termed the 'critical atomic approach value' for the action. The 'critical atomic approach value' for the reaction of the direct formation of iodides has been arrived at from considerations elucidated in previous papers and the distance of the closest approach of atoms of iodine has been calculated with the help of the author's formula. It has been suggested from the consideration of the direction of diffusion in the solid state that diffusion precedes the chemical action processes.

In previous communications<sup>2-6</sup> attempts have been made by the author to give some significance of the distance of the closest approach of atoms in chemical reactions. The reactivity of metals with water,<sup>2,3</sup> the reactivity of elements with bromine<sup>6</sup> and the capability of the metals and metalloids for the formation of amalgams<sup>4</sup> have been shown to be dependent on the distance of the closest approach of atoms of the elements concerned. In all these reactions, it has been found that in order that an element can react with these substances viz. water, bromine, mercury, it should have for the distance of the closest approach of its atoms a certain limiting value; this value has been shown to represent the distance of the closest approach of atoms of bromine<sup>6</sup> and mercury<sup>7</sup> in case of the formation of bromides and of amalgams respectively. This limiting value may be termed 'critical atomic approach value' for the reaction. Thus, the 'critical atomic approach value' for the reaction of metals with water, it may be noted, has been shown to be  $3.00 \text{ \AA}^{\circ}$ ,<sup>2</sup> for the reaction of elements with bromine,  $1.73 \text{ \AA}^{\circ}$ <sup>6</sup> and lastly for the formation of amalgams with mercury  $2.5 \text{ \AA}^{\circ}$ .<sup>7</sup> The object of the present paper is to bring out the significance of the distance of the closest approach of atoms of the elements concerned in their direct reaction with iodine, to arrive at the probable 'critical atomic approach value' for the reaction and to show that the value so obtained represent the distance of the closest approach of atoms of iodine, with which the elements react in this specified action. Iodine has been found to react with a large number of elements directly and the following list includes all the elements which enter into direct action with iodine. The values\* for the

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\* The values for the distances of the closest approach of atoms are taken from Bragg and Bragg's X-ray and Crystal Structure, Fifth Edition p. 168.

distances of the closest approach of atoms of the elements are given within brackets:

Copper (2.54), Silver (2.876), Nickel (2.505)  
Iron (2.48), Gold (2.88), Calcium (3.93)  
Magnesium (3.22), Cadmium (2.96) Mercury (2.50),  
Aluminium (2.86), Iridium (2.690), Titanium (2.96),  
Germanium (2.43), Tin (2.80), Lead (3.48),  
Antimony (3.37), Bismuth (3.47), Chromium (2.508),  
Tungsten (2.726), Silicon (2.35), Cerium (3.64),  
Zirconium (3.18), Sodium (3.72), Potassium (4.50),  
Sulphur (—), Zinc (2.67), Phosphorus (—)  
Indium (3.24), Thorium (3.54), Cobalt (2.514).

It is evident from the above table, that in the case of elements which combine directly with iodine, the distance of the closest approach of their atoms appears to be greater than an approximate value  $2 \text{ \AA}$ .

Now, as has already been referred to it has been shown by the author that the distances of the closest approach of atoms of bromine and of mercury determine their capability of the formation of bromides and amalgams respectively. The distance of the closest approach of atoms of bromine namely,  $1.73 \text{ \AA}$ , is characteristic of bromide formation as those elements whose atomic approach values are greater than  $1.73 \text{ \AA}$  are capable of reacting with bromine directly. In the formation of amalgams also, the distance of the closest approach of atoms of mercury namely,  $2.5 \text{ \AA}$  is characteristic in-as-much as the elements which are capable of forming amalgams have their atomic approach values greater than  $2.5 \text{ \AA}$ . Arguing on the same grounds, we may likely draw the conclusion that the 'critical atomic approach value' for the reaction of the direct formation of iodides in all probability would approximate to  $2 \text{ \AA}$  in view of the fact that it is only those elements which have the values for the distance of the closest approach of their atoms above the approximate figure  $2 \text{ \AA}$  are capable of reacting with iodine directly, and that any element having atomic approach value lower than this approximate value, for instance, carbon having atomic approach value  $1.54 \text{ \AA}$  or  $1.50 \text{ \AA}$  has not been found to enter into any chemical reaction with it. Further, this approximate value

2 Å° for the critical atomic approach value for the action would probably represent the least distance between the atoms of iodine.

This conclusion, it should be noted, finds confirmation in the fact that the value for the distance of the closest approach of atoms of iodine as calculated from the author's <sup>7</sup> formula  $D = \frac{P}{V_i \times d^{k/v}}$  is 1.75 Å° (approximating to 2 Å°) which is intermediate between that of silicon, viz. 2.35 Å° with which it reacts directly and that of carbon viz. 1.54 Å° or 1.50 Å° with which it has not been found to enter into direct action.

Much like the rule of the reactivity of metals with water<sup>2</sup>, that of the formation of amalgams<sup>3</sup>, that of the formation of complex compounds<sup>5</sup> and that of the reactivity of elements with bromine<sup>6</sup> we may hence lay down a rule for the reactivity of the elements with iodine that it is only those elements which have their values for the distances of the closest approach of atoms above 1.75 Å° are only capable of entering into direct reaction with iodine.

#### CALCULATION OF THE DISTANCE OF THE CLOSEST APPROACH OF ATOMS OF IODINE

A formula has been proposed by the author<sup>7</sup> for the calculation of the distance of the closest approach of atoms of elements from parachor, ionisation potential, atomic diameter and valency of the elements concerned. The formula, it may be noted, gave values in case of about dozen elements for the distance of the closest approach of atoms in close agreement with the experimental ones. It seems now worth showing the calculation for iodine and the value so obtained may be, with some amount of certainty, taken to represent the actual value for the distance of the closest approach of atoms of iodine. The formula may be represented thus:

$$D = \frac{P}{V_i \times d^{k/v}}$$

where D—distance of the closest approach of atoms of the element in question

V<sub>i</sub>—ionisation potential

d—atomic diameter

v—valency

k—a constant having the value 1.58.

For iodine, we have,

Parachor ... 91.0<sup>9</sup>

Ionsation Potential. 10.2<sup>10</sup>

Atomic Diameter .. 2.80<sup>11</sup>

It would appear on calculation with the help of the above formula, the distance 'D' of the closest approach of atoms of iodine is 1.75 Å<sup>c</sup> which is intermediate between the value for silicon on the one hand and for carbon on the other.

We, therefore, arrive at the following conclusions :

(1) The 'critical atomic approach value' for the reaction of the direct formation of iodides is 1.75 Å<sup>c</sup>.

(2) This 'critical atomic approach value' for the action, viz. 1.75 Å<sup>c</sup> represents the distance of the closest approach of atoms of iodine.

(3) Elements having their values for the distances of the closest approach of their atoms above 1.75 Å<sup>c</sup>, the value for iodine, are capable of reacting with iodine directly.

The dependence of chemical reactivity on the distance of the closest approach of atoms is thus quite apparent. But the exact function played by the distance of the closest approach of atoms cannot, at present, be clearly brought out. An attempt may, however, be made to make a rough idea of the part played by the 'closest atomic approach value' in such chemical reactions e. g. reaction of the direct formation of bromide and iodides and that of the formation of amalgams. Such an idea can be gathered from a study of the phenomenon of diffusion of elements in the solid state.

In a previous communication<sup>5</sup> it has been shown by the author that the diffusion of elements in the solid state depends upon the distance of the closest approach of atoms of the element which diffuses as well as upon that of the element into the body of which it diffuses. The rule which has been enunciated is that in the process of diffusion taking place in the solid state it is the elements which have lower values for the

distance of the closest approach of their atoms that are drawn into the bodies of elements which have higher values for the closest atomic approach.

Now from what has been said previously the capability of an element to react with another seems to be guided by its having an atomic approach value lower than that of the element with which it would react. Thus in diffusion in the solid state as well as in chemical action, the fact is quite apparent that it is the element possessing the lower atomic approach value that is only capable of diffusing into or reacting with an element which possesses a higher value for the closest atomic approach. It may hence be suggested that diffusion precedes such chemical reaction process; it is only when the diffusion of the atoms of an element has taken place into the body of another element with which it would react that some extra-ordinary force may be brought to play which may effect the chemical union. In purely diffusion process not attended by chemical action, probably the extra-ordinary force is not called to play or if at all, it is not so much active. The suggestion is, however, speculative. Further elucidation of the problem will be taken up later.

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# RESPIRATORY RESPONSE OF RIPE TOMATOES AND DORMANT POTATOES FOLLOWING WOUNDING

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## SUMMARY

It has long been known that when a plant organ is wounded an increase in the respiration intensity results. This increase has been variously attributed to the escape of accumulated carbon dioxide, the irritable response of the protoplasm to the stimulus of the oxygen of the air and metabolic changes in the wounded tissues. Data obtained with regard to the internal carbon dioxide concentration and the respiration rates of whole and wounded fruits and tubers indicate that the increased respiration following injury is referable in the main to the leakage of carbon dioxide through the wounds.

Although it has been known for a considerable time that wounded plant organs respire more intensely than those which are intact, there appears to be no unanimity among the various investigators with regard to the causes of this increased respiratory activity. For example, Magness<sup>2</sup> explained the rise in the respiratory activity of potatoes on the assumption that wounds facilitate gaseous exchange leading to a sudden escape of accumulated carbon dioxide. Lutman<sup>6</sup> believed that the increased respiration due to injury in the potato may be regarded, in the main, as an irritable response of the protoplasm to the stimulus of the contact of the tuber cells with the oxygen of the air. Gruss<sup>3</sup> found in the cells around wounds in potatoes an accumulation of sugars in the subphellogen and an increase in diastatic and oxidase activities. Friedrich<sup>3</sup> noted in the cells bordering the cut surfaces in potato, an increase in the total nitrogen, acidity and reducing sugars. It was observed by Hopkins<sup>5</sup> that the sugar content of wounded potatoes increased from 53 to 68 per cent of the original amount. In the light of the above-mentioned researches, it would be interesting to know to what extent increased respiration following wounding is due to mechanically facilitating gaseous exchange and to what extent it is due to actual metabolic changes in the wounded tissues. To elucidate this point ripening tomatoes (Sutton's Abundance) and dormant potatoes (Farrukhabad) were chosen for experimentation.

TOMATO (*Lycopersicum esculentum*)

The variety Abundance is preferable on account of the fact that all mature fruits are spheroidal in shape and tomatoes possessing approximately equal weights are easily selected. In view of the researches of Blackman and Parija<sup>1</sup>



it is imperative that the fruits grouped under one lot for experimentation should be of more or less the same metabolic age. Gustafson<sup>4</sup> has shown that during the ripening of tomatoes the colour of the fruit provides a general indication of the physiological age of the fruit. On the basis of colour ripening tomatoes were segregated into eight lots, each of which was divided into two sub-lots. One sub-lot was used for the determination of dissolved CO<sub>2</sub> which was estimated after the manner\*\* described by the authors elsewhere.<sup>9</sup> For the determination of respiration single fruits were used and the measurements were made with Singh and Mathur's modification of Dixon's respirometer<sup>8</sup>, larger respiration flasks being used than those recommended for seeds. After the respiration of a single fruit had been recorded, the skin of the fruit was removed from an area  $\frac{3}{8}$ "  $\times$   $\frac{3}{8}$ " by means of a sharp scalpel. After the determination of the rate of respiration, a second wound was inflicted involving the same area in an antipodal position and again the CO<sub>2</sub> evolution determined. Relevant data are recorded in Table I.

Table I

Dissolved carbon dioxide and respiration rates of whole and wounded tomatoes\*  
Temperature  $30 \pm 0.1^\circ\text{C}$ .

Dissolved CO <sub>2</sub> ml. per kilo	Respiration of the fruits			CO <sub>2</sub> due to	
	whole	with one wound	with two wounds	first wound	second wound
	ml. per kilo per hour				
19.9	17.71	22.11	22.19	4.40	0.08
20.6	19.31	24.42	24.52	5.11	0.10
21.3	20.31	25.98	26.09	5.67	0.11
22.0	23.70	29.99	30.12	6.29	0.13
26.9	30.31	38.21	38.39	7.90	0.18
29.8	29.37	38.42	38.80	9.21	0.38
30.2	26.29	36.31	36.80	10.09	0.49
35.0	20.11	30.41	30.92	10.39	0.51

\*All figures given above represent averages of 10 determinations.

\*\*The method consists, in brief, in submerging the plant material in boiling 90% alcohol in a closed container connected with two absorption towers (0.2N Na OH) which are in communication with an aspirator. The amount of dissolved carbon dioxide is estimated by titration with standard acid,

An examination of the data indicates that the dissolved  $\text{CO}_2$  progressively increases during the ripening of the tomatoes, a fact to which attention has already been called by the authors <sup>9,10</sup>, and that there is a parallelism between the values of dissolved  $\text{CO}_2$  and the extra amounts of  $\text{CO}_2$  liberated due to the wounds. It is striking to note that the extra amount of  $\text{CO}_2$  liberated due to the first wound is much greater than that evolved due to the second injury.

POTATO (*Solanum tuberosum*)

The first experiments were conducted on tubers that had been in storage at  $25^\circ \text{C}$ . for about  $1\frac{1}{2}$  months, and at intervals of a week thereafter. In all cases round potatoes of approximately the same size were selected for respiration and dissolved  $\text{CO}_2$  determinations. The manner of inflicting the wound was essentially the same as in the case of the tomato. The data (Table II) are similar to those

Table II

Dissolved carbon dioxide and respiration rates of whole and wounded potatoes \*  
Temperature  $30 \pm 0.1^\circ \text{C}$ .

Dissolved $\text{CO}_2$ ml. per kilo	Respiration of the tubers			$\text{CO}_2$ due to	
	whole	with one wound	with two wounds	first wound	second wound
	ml. per kilo per hour				
9.2	7.93	8.89	8.94	0.96	0.05
10.1	7.01	8.00	8.07	0.99	0.07
11.7	6.13	7.12	7.19	0.99	0.07
11.9	5.59	6.62	6.71	1.03	0.09
12.7	4.29	5.65	5.78	1.36	0.13
13.9	4.36	5.76	5.90	1.40	0.14
15.2	3.73	5.51	5.67	1.78	0.16
16.2	3.22	5.01	5.18	1.79	0.17
16.7	3.17	5.00	5.17	1.83	0.17
17.3	2.39	4.36	4.55	1.97	0.19

\*All figures given above represent averages of 10 determinations.

obtained for tomato, the extra amounts of  $\text{CO}_2$  liberated due to the wounds being positively correlated with the values for dissolved carbon dioxide. This is also discernable that the extra amounts of  $\text{CO}_2$  liberated due to the first wounds are in all cases much greater than those due to the second ones.

#### DISCUSSION

If metabolic changes in the wounded tissues are the cause of increased respiration one is justified in believing that the extra amounts of  $\text{CO}_2$  evolved due to injuries of an equal magnitude will be more or less equal when the two determinations are made within a few hours of each other. Although the data do not justify the suggestion—it appears highly probable—that in wounded ripe tomatoes and injured dormant potatoes the extra  $\text{CO}_2$  evolved due to metabolic changes is negligible in comparison with that liberated due to the facilitation of gaseous exchange occasioned by the injury, they do indicate that most observations on increased respiration concomitant upon injury are probably referable, in the main, to a leakage of  $\text{CO}_2$  through the wounded portions.

Another point of considerable interest is the disparity between the respiration rates of whole fruits and tubers and their internal  $\text{CO}_2$  concentrations (Tables I and II). In a recent note<sup>10</sup> from this laboratory it was shown that the permeability of the superficial tissues to the diffusion of gases is a factor determining the amount of  $\text{CO}_2$  evolution from ripe tomatoes. A decreased permeability of the external tissues naturally gives rise to a lessened rate of respiration as measured by the rate of superficial evolution of carbon dioxide. To determine the permeability of the periderm of potato, the gas-extraction apparatus used by Smith<sup>11</sup> was employed. Ten potatoes, approximately alike in size and shape, were selected for each determination, the gas being collected in a burette after three minutes' extraction in a Torricellian vacuum. Amounts of gas extracted from 10 potatoes at successive weekly intervals during the storage period were as follows: 6.25, 5.70, 5.20, 4.95, 4.65, 4.05, 3.70, 3.20, 2.95, and 2.75 ml. A perusal of the data indicates that the permeability of the periderm considerably decreases during the storage life of potatoes, a striking parallelism being obtained between the values of gas extracted from the potatoes and their respiration rates. This indicates that the superficial evolution of  $\text{CO}_2$  under various circumstances will be conditioned, among other factors, by (1) the concentration of  $\text{CO}_2$  inside the tuber and (2) the permeability of periderm to diffusion of gases. In view of

the fact that the true measure of "respiration" is the rate at which the substrate is broken down to simpler substances, it is obvious that the estimation of respiration based upon the rate of  $\text{CO}_2$  evolution, particularly in storage organs, may not give a true measure of the rate of cellular respiration.

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THE MICROSCOPIC ANATOMY AND HOMOLOGY OF THE GLANS AND  
THE OS PENIS OF THE COMMON INDIAN GROUNDSQUIRREL  
(*FUNAMBULUS PALMARUM*)

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SUMMARY

A complete series of both transverse and sagittal sections was prepared of the terminal end of the penis of the common Indian Ground-squirrel (*Funambulus palmarum*). The os penis and the glans have been described and homologised with the penis in man.

The penis of the members of the family Sciuridae shows a variety of forms. The accompanying fig. 1 shows the penis of Fox squirrel, Flickertail Gopher, Chipmunk, Red squirrel, Flying squirrel, *Spermophile* and the Indian ground squirrel. Although they differ considerably from each other even as seen with the naked eye, yet they have certain features in common. For example all of them have a marked terminal, ventral bend and the anterior ends of the crura are pointed and ossified.

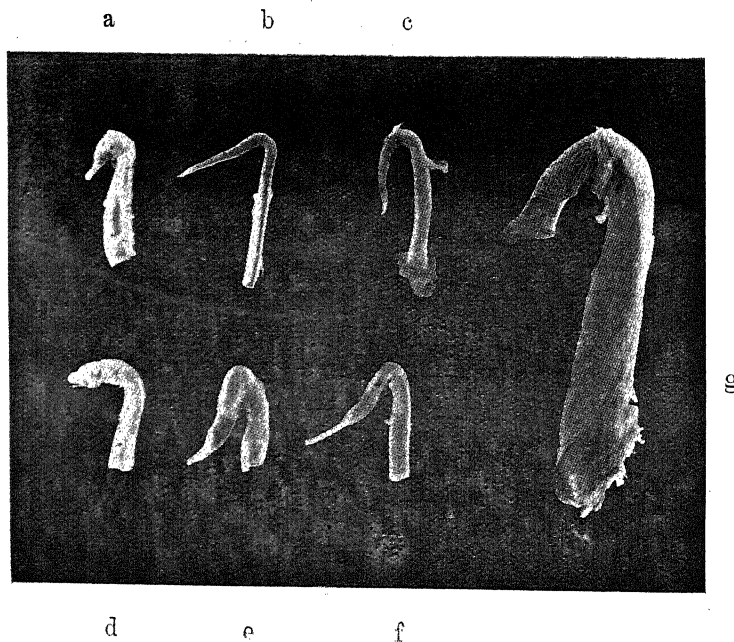


Fig. 1

Terminal end of the penis of Sciuridae. a, Flicker tail Gopher (*Citellus richardsoni*); b, Red squirrel (*Tamiasciurus hudsonicus*); c, Chipmunk (*Tamias striatus*); d, *Spermophile*

(*Citellus tridecemlineatus*); e, Indian Ground squirrel (*Funambulus palmarum*); f, Flying squirrel (*Glaucomys volans*); g, Fox squirrel (*Sciurus niger*).

The main difference which is responsible for the variety in forms of the organ concerns the extent and size of the anterior end of the crura and the glans penis, which differ from member to member. This paper is based on complete transverse and sagittal serial sections of the terminal end of the penis of the common Indian Groundsquirrel (*Funambulus palmarum*). This animal is found abundantly in a wild state throughout the Gangetic plain in India and a few of them were trapped for this investigation. The tissue was decalcified in a solution of 3% nitric acid in 70% alcohol and later imbedded in celloidin. Haematoxylin and eosin were used as stains for the sections.

Fig. 2 is a photograph of the terminal end of the penis of the animal. The crura bends ventralwards at a sharp acute angle and disappears into the substance of the softer reddish brown oblong swelling of the glans.

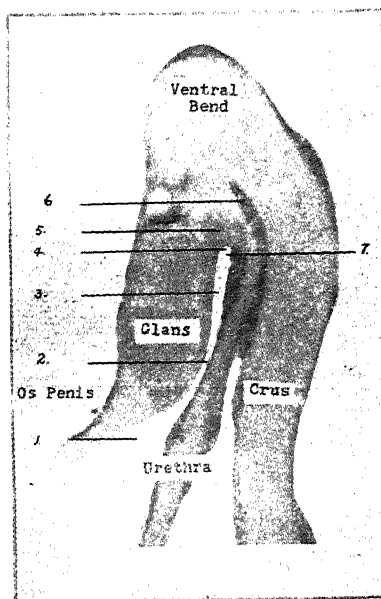


Fig. 2

Photograph of the terminal end of the penis of the common Indian ground squirrel (*Funambulus palmarum*) which was serially sectioned. Numbered cross lines indicate the levels at which sections have been microphotographed for the text.

The Os penis which is the terminal ossified end of the crura emerges from the distal end of the glans as a beak-shaped pale white process. It is smooth

and shiny and feels bony to the touch, although on a microscopic examination it is seen to be covered by a thin layer of cavernous tissue which has been evaginated out from the glans along with the urethra.

The urethra surrounded by the corpus cavernosum is lodged in the ventral crural groove proximal to the ventral bend and easily strips away like a string. Distal to the bend however, it runs through the substance of the glans in close proximity to the caudal aspect of the Os penis as seen in fig. 3. Distal to the glandular swelling it runs close to the Os penis and winds round the bone to open near its tip on its ventral aspect.

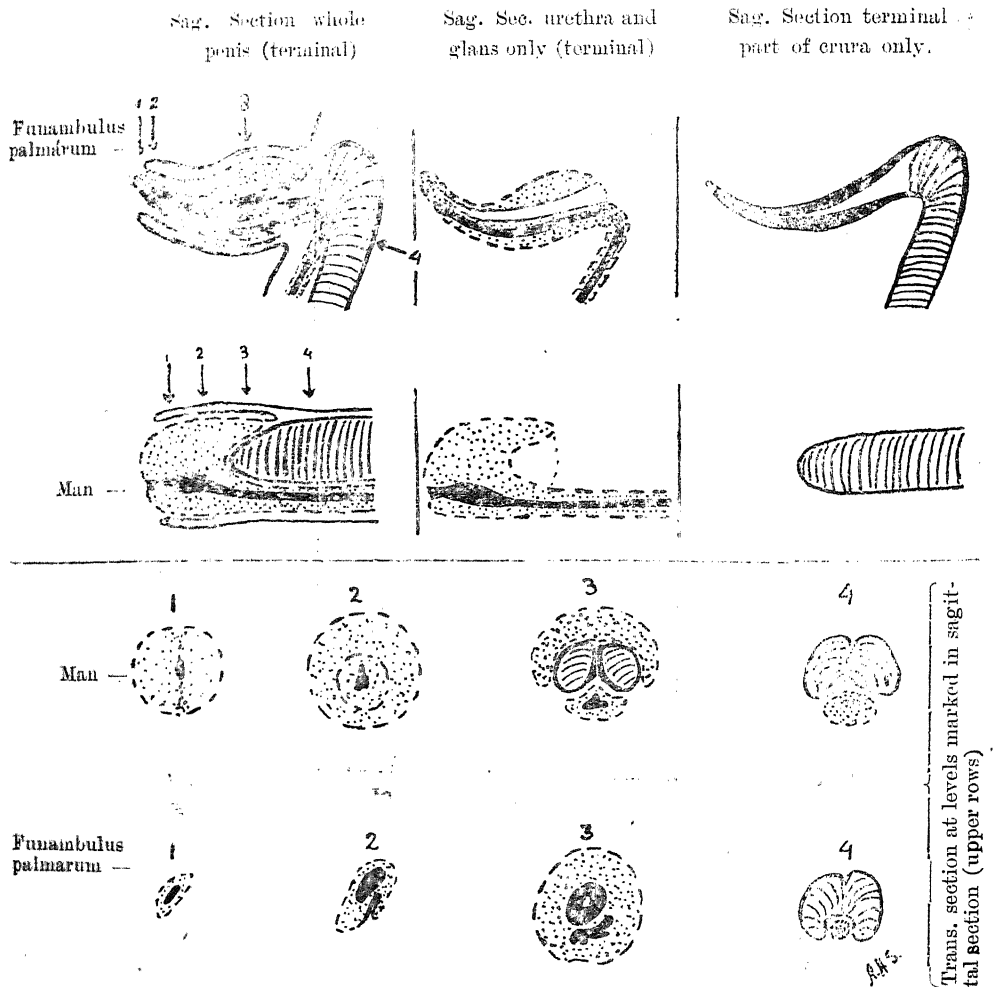


Fig. 3

The above diagram homologises the structures composing the terminal end of the penis of *Funambulus palmarum* and the penis of man. It will be noticed that the general construction and relations are the same. There are two characteristic features which distinguish the crura in *Funambulus palmarum*.

1. The terminal end instead of ending bluntly as in man into the posterior part of the glans near corona glandis is beak shaped and extends much farther forwards carrying a thin covering of glandular tissue over itself.

2. The beak shaped terminal end of the crura is bony and is composed of lamellae round a central canal.

Fig. 4 *h* shows the urethra winding by the side of the Os penis from the dorsal aspect to reach its orifice on the ventral concave aspect as seen in fig. 4 *i*

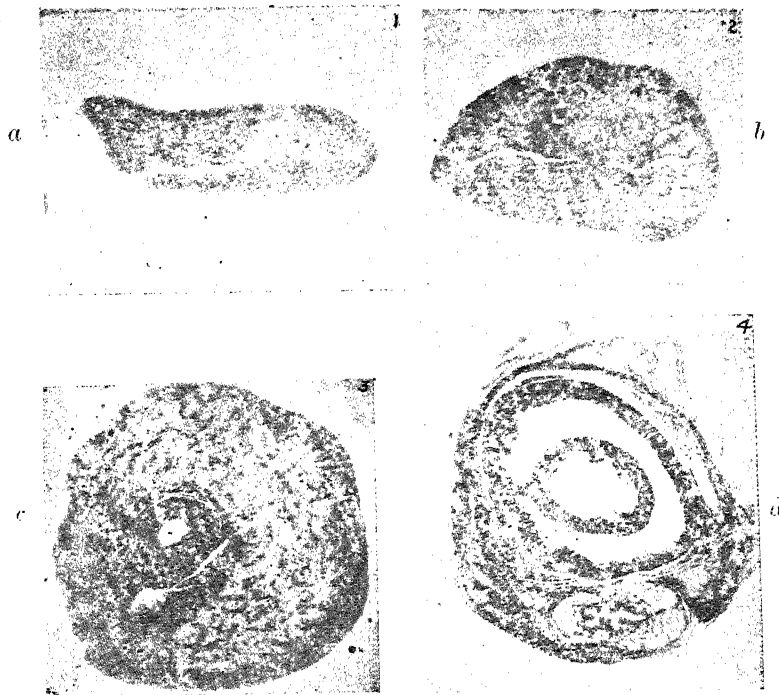


Fig. 4 (*a-d*)



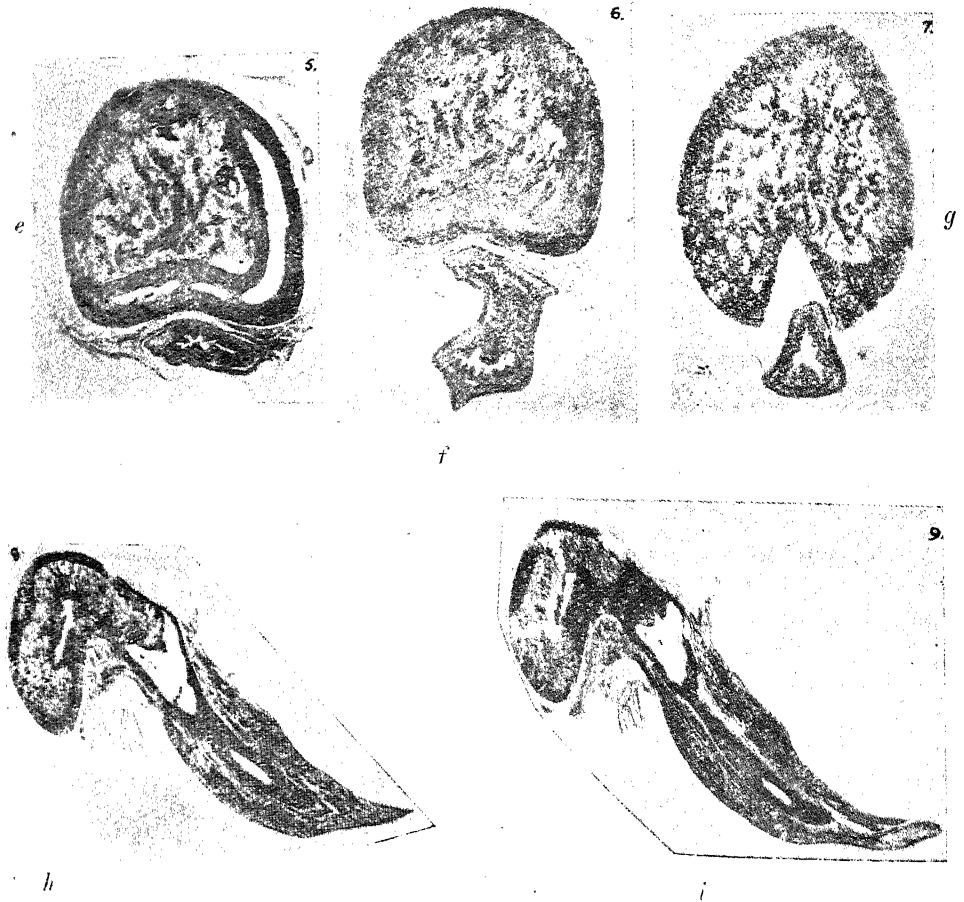


Fig. 4 (e-l)

*a.* Section of tip of Os Penis passing through external orifice of urethra. The Os Penis is surrounded on all sides by cavernous tissue which it has evaginated forwards from the glans.

*b.* Section through distal end of glans. The Os Penis shows the lamellar arrangement around a central canal.

*c.* Section through the middle of the glans.

*d.* Section near the Corona glandis. It shows the corpus cavernosum urethrae surrounding the urethra and the semilunar tissue of the glans surrounding the base of the Os Penis. The canal of the bone is very wide at its base.

*e.* Section proximal to corona glandis. It shows the muscular crura and part of the base of the Os Penis in direct continuity.

*f.* Section through the ventral bend of the urethra. It shows the muscular crura and its ossification.

g. Section of the Penis proximal to the ventral bend. It shows both sides of the crura fused into a common mass and presenting a ventral groove for the lodgment of the urethra surrounded by its own sheath.

h & i. Sagittal sections of the terminal end of the Penis. These show the muscular crura directly continuous into the Os Penis. The central canal of the Os Penis is wide at the base and gradually narrows down towards the apex of the bone. The urethra running through the ventral aspect of the substance of the glans gains the dorsal concave side near the tip.

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STUDIES ON THE FAMILY DIPLOSTOMIDAE, POIRIER.  
PART I—TWO NEW PARASITES OF THE GENUS *DIPLOSTOMUM*  
V. NORDMANN FROM INDIAN CARNIVOROUS BIRDS

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SUMMARY

A brief historical account of the genus *Diplostomum* V Nordmann is given. Two new species namely, *Diplostomum ketupanensis* n. sp. and *D. buteli* n. sp. are described. The species are very remarkable in having A-shaped or slightly H-shaped testes—a feature which readily distinguishes them from all the hitherto known species.

GENUS: *DIPLOSTOMUM* V. NORDM., 1832.

(Syn. *Proalaria* La Rue, 1926.)

Large numbers of hemistomes, many of which were restudied by Krause in 1915 have recently been allocated to different genera. La Rue (1926) created the genus *Proalaria* under the sub-family *Polycotylinæ*, for forms characterised by the distinct division of body into fore and hind parts, possession of lateral suctorial cups and circular or elliptical holdfast organ with or without cavity. He assigned the following species to this genus:—*Proalaria spathaceum* (Rudolphi) *P. gariae* (Guberlet), *P. indistincta* (Guberlet), *P. confusa* (Krause), *P. excavata* (Rud), *P. triangular* (Johnston), *P. intermedium* (Johnston), *P. triloba* (Rud), and *P. huronensis* (La Rue). A year later Dubois described three species as *Hemistomum colymbi*, *H. pusillum* and *H. glossoides* of which the last has been transferred as the type species of a new genus *Glossodiplostomum* Dubois 1932. He drops the genus *Proalaria* and considers it to be synonymous with *Diplostomum* on the basis of priority. Ciurea (1928) and Cort and Brooks (1928) have described *D. clavatum* and *D. hexicaudatum* respectively. Chandler (1932) reported *D. variabilis* from

the opossum, *Didelphys virginiana*. *Diplostomum butasturina* from *Belosiar indicus*, described by Tubangui (1932), is considered by Dubois (1936) as *Neodiplostomum butasturinum*. Recently a number of species have been added to the genus, namely *D. mergi* Dubois (1931), *D. parvicerosum* Dubois (1931), *D. pelmatoides* Dubois (1931) *D. orientale* Yamaguti (1933), *D. vanelli* Yamaguti (1935) and *D. alcedense* Patwardhan (1935). The species, *D. confusum* has now been transferred as the type species of a new genus *Bolbophores*, set up by Dubois (1935). In this paper are added two more remarkable species belonging to this genus.

*Diplostomum ketupanensis*, n. sp.

About two dozen specimens were found parasitic in the small intestine of the northern brown fishing owl, *Ketupa zelonensis handericki*, examined in September 1936. Out of a dozen specimens of the host examined, only three have been found infected. The parasite occurs in association with two interesting trematodes of the genera, *Neodiplostomum* and *Striega*. The translucent white gravid worms, 1.88-3.2\* in length and flexed dorsal at the junction of the two body regions, have a body distinctly divided into fore and hind parts. The spoon-shaped fore body with foliate lateral edges united posteriorly, measures 0.992-1.52 in length and 0.88-1.04 in maximum width, which lies immediately in front of the holdfast organ, while the hindbody which arises from the dorsal side of the forebody, is cylindrical, 0.896-1.68 in length and 0.528-0.704 in width and rounded at the posterior extremity.

The oral sucker,  $0.08 \times 0.118-0.128$  in size, is terminal and somewhat longer than broad. It opens into a small narrow prepharynx, 0.018 in length; the pharynx is small and oval, 0.048-0.054 in transverse diameter. The short and narrow oesophagus bifurcates into two long slender caeca, which run through the glandular anterior part of the forebody and passing on the dorso-lateral side of the acetabulum and the holdfast organ, extend into the hindbody running laterally and ventrally to the gonads, and terminating blindly near the posterior extremity. The prominent ear like pseudo-suckers are 0.16 long and 0.064 broad. The numerous muscle fibres, arising from the basal portion of the suctorial pockets, run backwards in two, dorsal and ventral, sets; the fibres run closely parallel to one another and seem to be attached to the dorsal wall of the posterior extremity of the hindbody. The transversely oblong acetabulum  $0.112 \times 0.144-0.16$ , lies 0.624 from the anterior extremity, a little posterior to the equator of the forebody, and distinctly separated from the holdfast organ

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\* All measurements are in mm.

by a distance of 0.096. The holdfast organ is somewhat longitudinally elongated and elevated into a prominent cone, specially when the body is flexed, measuring  $0.24-0.288 \times 0.192-0.208$  in size. It has an irregular median slit lined with thick cuticle. The adhesive gland is situated at its base.

The excretory system has not been worked out in detail.

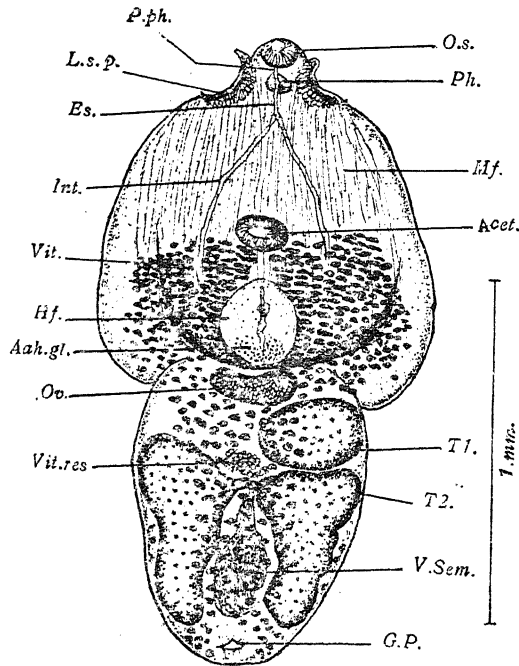


Fig. 1

*Diplostomum ketupanensis* n. sp. Ventral View.

acet. acetabulum; ov. ovary; adh. gl. adhesive gland; ph. pharynx; es. esophagus; p. ph. prepharynx; g. p. genital pore; t<sub>1</sub>. anterior testis; hf. holdfast organ; t<sub>2</sub>. posterior testis; l. s. p. lateral suckorial packet; v. sem. vesicula seminalis; m. f. muscle fibre; vit. vitelline follicles; o. s. oral sucker vit. res. vitelline reservoir.

The gonads practically occupy the anterior nine-tenth part of the hind body. The approximately wedge shaped, asymmetrical anterior testis of 0.352

0.384 length and 0.352-0.448 width lies to the left side of the median line in front of the equator of the hindbody. The posterior testis is considerably more bulky than the anterior one and practically occupies the whole width of the hindbody. It is more or less symmetrical H or better  $\Lambda$ -shaped with thick limbs. The latter, 0.64-0.992 long and 0.224-0.288 broad, are connected by a narrow isthmus, 0.032-0.048 in antero-posterior diameter. Between the two limbs lie the coiled vesicula seminalis, the descending limb of the uterus and the vas deferens. The anterior vas-efferens arises from the anteroventral side of the attenuated end of the anterior testis. After running anteriorad for a short distance, it turns backwards to join its fellow which arises from the mid ventral side of the narrow isthmus between the two limbs of the posterior testis. The vas deferens, thrown into sinuous curves, runs mesially to the voluminous, S-shaped vesicula seminalis which is situated somewhat dorsally. The goblet-shaped proximal portion of the latter, 0.2 in length and 0.112 in width, occupies nearly the whole of the space between the two limbs of the hinder testis.

The transversely oval, median ovary, 0.08-0.112 long and 0.288-0.352 broad, lies just near the body constriction. The oviduct arises from its postero-dorsal side and after giving off the Laurer's canal, runs posteriorad making a curve on the right side of the attenuated end of the anterior testis. The Mehlis gland is situated towards the right side of the median line, facing the inner end of the anterior testis. The uterus extends anteriorad to the level of the ovary and in its backward course as a sinuous descending limb, passes ventrally to the gonads. Its distal portion unites with the ejaculatory duct and opens through a small genital cone into the genital atrium. The latter communicates to the exterior, through the dorsally situated sub terminal genital pore. The uterine eggs are absent.

The vitellaria, consisting of closely massed follicles, extend anteriorly as far as the acetabulum, and posteriorly to the extreme hinder end of the body; they are broadest at the level of the holdfast organ. In the hindbody the main bulk lies ventrally in the pre and post testicular regions and only as a fairly wide median longitudinal strip in the testicular region. The vitelline reservoir is large and lies between the two testes, dorsal to the uterus and the vas deferens.

*Discussion* :— *Diptostomum ketupanensis* n. sp. can be easily distinguished from all the hitherto described species of the genus, on account of the

peculiar shape of its posterior testis, disposition, shape, and size of the vesicula seminalis, shape of the pseudosuckers, and the much greater development of the muscle fibres which traverse the parenchyma of the fore and hind parts.

*Diplostomum butei* n. sp.

A number of specimens were obtained from the small intestine of *Buteo rufinus rufinus* in September 1936. The feebly muscular dumb-bell shaped body, 1.052-1.118 in length, is sharply divided into the scoop shaped forebody and the cylindrical posteriorly rounded hindbody. The forebody is 0.467-0.551 long and 0.535-0.75 broad, while the hindbody is 0.567-0.585 long and 0.5-0.584 broad across the anterior testis. The foliate lateral margins of the forebody are much incurved and united behind the holdfast organ. The ratio in the length of fore and hind parts is approximately 4 : 5.

The subterminal, spherical, oral sucker measures 0.057-0.067 in diameter. The long barrel shaped pharynx is  $0.062 \times 0.034$ -0.039 in size. The prepharynx is very small ; visible in sections only. The oesophagus is short, 0.023 in length and the slender caeca extend to near the posterior end of the body. The lateral suctorial pockets are 0.034-0.05 long and 0.056 thick. The numerous muscle bands arising from these pockets run backwards on the dorsal side and are attached to the dorsal wall of the hindbody at or near the posterior end. The transversely oval and pre-equatorial acetabulum,  $0.023 \times 0.07$  in size, lies median close in front of the holdfast organ and 0.2 behind the anterior extremity. The large transversely oval to spherical holdfast organ,  $0.2$ - $0.25 \times 0.2$ - $0.3$ , has an irregular median cleft on the ventral surface. The adhesive gland with a posterior median incision, lies at the postero-dorsal part of the holdfast organ as a conspicuous mass of nuclei.

The gonads occupy the entire hindbody. The two testes with a marked concavity on the ventral side lie close behind one another. The asymmetrical anterior testis,  $0.15$ - $0.2 \times 0.417$ - $0.434$ , lies in the anterior third of the hindbody. The posterior testis is symmetrical H or  $\wedge$ -shaped in from and occupies the entire width of the hindbody. Its two limbs, approximately equal in size, 0.35 long and 0.184 broad are joined by a narrow bridge. The feebly developed vesicula seminalis lies immediately behind the second testis.

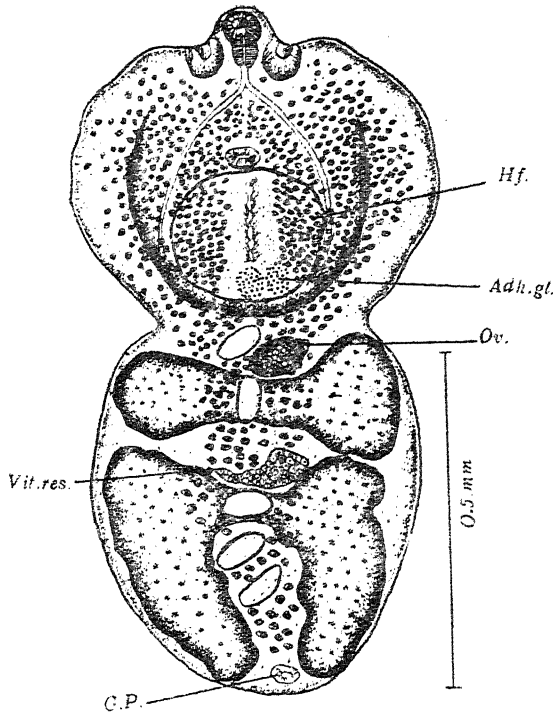


Fig. 2

*Diplostomum buteli* n. sp. Ventral view. Lettering as in Fig. 1

The sub-globular ovary,  $0.00-0.089$  in length and  $0.117$  in width, lies close in front of the anterior testis, near the body constriction, a little to one side of the median line. The shell gland complex lies close behind the inner attenuated end of the anterior testis. The uterus extends anteriorly to near the body constriction; its distal end along with the terminal portion of the ejaculatory duct opens into the spacious genital atrium with a dorsal and subterminal genital pore. The large yellow, operculate ova measure  $0.1 \times 0.042$  in size. The vitellaria which are profusely developed both in the fore- and hind parts, extend from the level of the pharynx to the sub-caudal region. In the forebody they occupy the entire width and are more concentrated in the lateral and adjacent parts of the holdfast organ. In the hindbody they form a longitudinal strip, situated mid-ventral to the gonads. The large vitelline reservoir,  $0.2 \times 0.084$  in size, lies between the testes.



*Discussion* :—Of all the species hitherto known, the new species resembles *D. ketupanensis* n. sp. in the form of its posterior testis, which is its most remarkable feature. But it differs from it, in the form and size of the body size of the suckers, relative size of the holdfast organ, the form and the size of the ovary and the anterior testis, shape and the disposition of the vesicula seminalis, shape of the pseudosuckers and above all the extent of the vitellaria.

I am very grateful to Dr. H. R. Mehra, under whose supervision this work has been done, for his valuable help and guidance. Thanks are also due to Dr. D. R. Bhattacharya for facilities in the Department.

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# ON THE AMYLASE FROM THE INDIAN WATER CHESTNUT

(*TRAPA BISPINOSA*, ROXB.)

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## SUMMARY

An active diastatic enzyme is found to be present in the fruits of *Trapa bispinosa* Roxb. The reducing power of the sugar formed was estimated by the method of Willstätter and Schudel. The reaction was found to follow a monomolecular course till 50-60% of the substrate is decomposed. The optimum temperature of the amylase is 50-55°C; and the optimum pH 5.2-5.6. The amylase is mainly a saccharogenic  $\beta$ -amylase in nature.

*Trapa bispinosa* Roxb., called 'Singhara' in Hindustani; 'Paniphal' in Bengali and 'Sringataka' in Sanskrit is an aquatic plant much cultivated in fresh water tanks or ponds throughout India. It belongs to the natural order Onagraceae. The fruits are 1 or 1½ inch long and broad with two apposite angles and with a scabrous spine. The white kernel contained within the red brown fruit is used as a food by large communities of the poorer classes in Northern India, and by those who keep fast on ceremonial occasions. The nuts are farminacious and considered by Indians as cool and sweet, useful in bilious affections and diarrhoea. The nuts are also used in the Punjab in the form of poultice.<sup>5</sup>

The present investigation relates to the presence of an enzyme the purification and characterisation of which has been described in the experimental portion of the paper. The optimum pH and the optimum temperature have also been determined and the effect of dialysis studied.

## EXPERIMENTAL.

*Preparation of the enzyme* :—A fresh supply of the fruits of *Trapa bispinosa* Roxb. was obtained from the neighbourhood of Allahabad, dried in the shade and finely powdered. After repeated trials it was found that finely powdered dried kernels of the fruit passed through a fine sieve and finally

through muslin can be preserved well in Jena glass bottles without any appreciable loss of the amylolytic activity. For all experiments freshly prepared enzyme solution was used by adding a known volume of water to a known weight of the powdered kernels.

*Method of procedure:*—Soluble starch prepared according to the method of Litner (Practical Organic and Bio-chemistry: Plimer 1931) was used as a substrate. The hydrogen ion concentration of the reaction mixture was adjusted by addition of Walpole's acetate buffer (pH 3.6-5.6) and for higher ranges of pH Sorensen's phosphate buffer was used (pH 5.6-8.0). The enzyme solution was obtained by taking a weighed amount of the enzyme powder, which was made into a thin paste with water and made upto 100 c. c in a measuring flask. It was prepared afresh for each experiment. The reactions were carried out in conical flasks at constant temperature in an electrically regulated incubator.

The digestions were carried out with 100 c. c lots of mixtures containing 1.5 gms of starch, buffer solution and enzyme solution. Aliquots of 10 c. c were drawn out at intervals of 15, 30, 45 and 60 minutes and the reducing power of the fraction estimated by the method of Willstätter and Schudel.<sup>9</sup> The degree of hydrolysis is expressed by the reducing power of the mixture being measured by the amount of the maltose formed in a given time. The velocity constants for a unimolecular reaction were calculated according to Euler and Svanberg<sup>1</sup> by applying the usual formula

$$K = \frac{1}{t} \log \frac{a}{a-x}$$

where K is the velocity constant; t, the time in minutes; a, the original amount of the substrate and x, the amount of the substrate decomposed in time t. It was found with my work that the reaction follows a monomolecular course till 50-60% of the substrate is decomposed. The activity of the amylase is given in terms of the velocity constants K, and the activity unit in terms of Eulers' units i.e.

$$Sf = \frac{K \times \text{maltose formed (in grams)}}{\text{Enzyme preparation (in grams)}}$$

where K is the velocity constant of the unimolecular reaction.

*Dialysis:*—100 grams of the powdered kernels of the fruit were treated with 500 c. c of distilled water in presence of toluene for about 24 hours at the room temperature (30-32°). It was then filtered first through muslin and then through

paper, the sediment was allowed to settle and again filtered. It was then dialysed in collodion bags against distilled water containing a little toluene the water being renewed twice every day. Each bag contained 50 c.c. of the enzyme extract. At definite intervals of time one bag was removed and the contents diluted to 100 c.c. The activity of this diluted dialysed enzyme solution was then estimated. The values thus obtained were compared with those of the specimens of the original enzyme extract which had been left undialysed.

For each experiment the reaction mixture consisted of  $75\frac{1}{2}$  of a  $2\frac{1}{2}\%$  starch solution, 20 c.c. of acetate buffer of pH 5.6, and 5 c.c. of enzyme solution. The temperature was  $30^\circ$  adjusted in a thermostat. The results are given in Table I

Table I

Dialysed for hours	Dry wt. enzyme solution (gm/5 c.c)	Activity K $\times 10^4$	Activity Unit Sf at $30^\circ\text{C}$
0	0.0500	61.2	0.129
20	0.0090	45.9	1.152
40	0.0085	25.6	1.260
60	0.0072	12.5	1.024
80	0.0066	10.2	0.920
100	0.0051	8.4	0.620
120	0.0051	6.0	0.502

The results obtained show that the activity of the enzyme per unit weight of the dried substance i.e. Sf increases after 20 to 40 hours of dialysis to nearly ten times. It seems evident, therefore that dialysis can very successfully be applied as a means of purifying the enzyme extract.

These observation are in harmony with those of Euler and Svanberg<sup>2</sup> and those of Holmberg<sup>1</sup> who used dialysis for the purification of malt and liver amylases and got preparations of highly increased enzyme activity. Giri<sup>3</sup> also showed that the activity of sweet potato amylase is increased seven times on dialysis for 16 to 40 hours.

*Effect of hydrogen ion concentration on the activity of amylase*:—For these experiments Erlenmeyer flasks containing 75 c.c of a 2% starch solution, 20 c.c of buffer solution of increasing hydrogen ion concentrations were kept in a thermostat adjusted to 35° C. When the reaction mixture had attained the temperature of the thermostat, 5 c.c of the enzyme solution prepared by dissolving 5 grams of the powdered kernels in 100 c.c of distilled water were added and the activity measured in each case by drawing out 10 c.c of the mixture. Control experiments were done, using 5 c.c of distilled water in place of enzyme solution, in order to eliminate any error. The results are given in Table II below.

Table II

Starch concentration 1.5% ; enzyme concentration 0.25% ; temperature 35°C.

pH	Activity ( $K \times 10^4$ )
3.6	20.8
4.0	39.9
4.4	65.1
4.8	75.7
5.2	96.9
5.6	97.2
6.0	90.0
6.5	70.2
6.8	54.8
7.2	45.9

It will be evident from the table that the amylase acts best in the region of pH 5.2 to 5.6. These results are quite in harmony with other amylases of vegetable origin, which had their optimum pH between 5 and 6.

*Effect of temperature on the activity of the amylase*:—The reactions were carried out in conical flasks containing 75 c.c of a 2% starch solution, 20 c.c of the buffer solution of pH 5.6 (Walpole's acetate buffer) and 5 c.c of the enzyme solution of the same concentration as above. The flasks were maintained at different temperatures as shown in the table below.

and the activity determined by estimating the amount of maltose formed in a given time. Control experiments were always done as in the previous case in order to minimise any error that may creep in. The results are given in Table III.

Table III

Starch concentration 1.5%; enzyme concentration 0.25%; pH 5.6.

Temperature °C	Kx 10 <sup>4</sup> (activity)
20	40.2
25	53.2
30	61.0
35	78.0
40	93.0
45	106.9
50	115.2
55	118.9
60	99.0
65	79.0
70	55.0

These results show that the amylase acts best in the region of temperature 50-55° c. In this connection it is very gratifying to note that the optimum temperature of amylases of germinating grains is 50-55° C. that of sweet potato is 50-56° C. In the case of amylases obtained from animal origins, however, the optimum temperature varies considerably and lies between 35° for pancreatic amylase to 45° for salivary amylase.

*The nature of the amylase* :—It has been observed by a series of workers that amylases can be divided into two classes viz.  $\alpha$ -or dextrinogen amylase and  $\beta$ -or saccharogen amylase. The former decompose starch only upto dextrins stage and the latter produce maltose from it. Thus Nordh and Ohlsson<sup>7</sup> and Myrback and Myrback<sup>8</sup> have shown that the amylase from ungerminated barley consists mostly of  $\beta$ -amylase, whereas Willstätter and Rohdewald<sup>9</sup> have shown that the

amylase of leucocytes are dextrogenic in character, thereby showing a close resemblance to pancreatic amylase and differing from malt amylase which is a pure form of  $\beta$  amylase.

The nature of *Trapa bispinosa* amylase was determined as follows : 25 c.c. of a 2% starch solution, 10 c.c. of Walpole acetate buffer of pH 5.6 were maintained at 35° C and 5 c.c. of the 5% enzyme solution added. After about one hour 10 c.c. of the reaction mixture were withdrawn and the reducing power determined. This gave a measure of the activity of  $\beta$ -amylase. The activity of  $\alpha$ -amylase was determined viscometrically. The results are given below :—

	Activity of the saccharogenic amylase	Activity of the dextrogenic amylase.
<i>Trapa bispinosa</i>		
amylase	40	10.5

These figures clearly show that the amylase is mainly a  $\beta$ -amylase, very little quantity of  $\alpha$ -amylase being present.

In conclusion the author begs to acknowledge his grateful thanks to Dr. S. Dutt D.Sc. for his keen interest in the work and to Prof. N. R. Dhar for encouragement.

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## SUPERPOSED PARASITISM OF *CUSCUTA REFLEXA* ROXB.

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### SUMMARY

During the course of investigations on the physiology and chemistry of phanerogamic parasitism an interesting case of double-parasitism in which *Cuscuta* was found to infect *Loranthus* when both of them were growing on a common host, *Bauhinia*, has been recorded. The capacity of the two parasites to grow harmoniously on a single host for long periods is probably explicable on the basis of the capacity of *Cuscuta* to adjust its osmotic and H-ion concentrations over a fairly long range. Data obtained on artificial injection of sugars indicate that *Cuscuta* can more efficiently absorb nourishment on account of its power of rapidly adjusting its sap concentration in accordance with the nutritional conditions of the host.

It is a matter of common observation that the majority of phanerogamic parasites attack angiospermic herbs, shrubs and trees, and sometimes also exhibit self-parasitism as in *Cuscutaceae* and *Lauraceae*. Kerner and Oliver<sup>3</sup> have reported instances of interspecific parasitism exhibited by *Loranthus buxifolius* upon *L. tetrandrus* in Chile, European mistletoes parasitising *Loranthus* in Verona, and *Viscum moniliforme* affecting another species *V. orientale* in India. No case of intergeneric parasitism seems to have been recorded amongst higher plants although good many instances are common in bacteria and fungi.

During the course of a detailed experimental study on the physiology and chemistry of phanerogamic parasitism conducted at this Experiment Station, there was discovered an interesting case of parasitism between two widely different genera *Cuscuta* and *Loranthus*. A completely leafless parasite like *Cuscuta reflexa* Roxb. was found to infect the partially-leafy *Loranthus longiflorus* Desv. when both of them were found to grow on a common host *Bauhinia purpurea* L. Fig. 1 shows the initial parasite *Loranthus* growing on this species and being attacked by *Cuscuta*. Quite a number of cases were discovered in which *Loranthus*, a stationary parasite, was found to confine itself to one branch of its host only, while *Cuscuta* due to its ever-spreading and



trailing habits, attacked gradually the adjacent branches as well as its fellow accomplice *Loranthus* if it came in contact with it.



Fig. 1

*Cuscuta* attacking *Loranthus* growing on *Bauhinia purpurea*

The peculiar behaviour of such phanerogamic parasites has been studied by Harris<sup>1</sup>, Harris and Lawrence<sup>2</sup>, Parija and Saran<sup>4</sup> besides others. It has been indicated that parasitism is dependent upon some constitutional differences in the cell sap characteristics of the host and the parasite. The parasites, it is observed, not only maintain higher sap concentrations but are

invariably capable of adjusting the same in due response to the changing concentrations of the host. *Cuscuta* being more plurivorous, appears to maintain a higher sap concentration over that of the host as well as *Loranthus* (Table I).

Table I

Variations in osmotic concentration and pH of the host and the parasites

Plant	Before <i>Cuscuta</i> attacked <i>Loranthus</i>		After <i>Cuscuta</i> attacked <i>Loranthus</i>			
	Os. con. in atmos.	pH.	By natural infection		By artificial infection	
			Os. con. in atmos.	pH.	Os. con. in atmos.	pH.
1. <i>Bauhinia purpurea</i> L. (common host).	13.27	5.8	13.26	5.8	13.26	5.8
2. <i>Loranthus longidorsus</i> Desv. (initial-parasite).	16.83	5.5	16.86	5.5	16.85	5.5
3. <i>Cuscuta reflexa</i> Roxb. (superposed parasite).	15.03	5.4	17.01	5.3	17.00	5.3

The variability of osmotic and H-ion concentration in *Cuscuta* from 15.03 to 17.01 atmospheres, and 5.4 to 5.3 pH respectively would suggest that this parasite changes its sap concentration over wider limits. A higher gradient is always maintained in *Cuscuta* as compared to *Loranthus* itself and the common host on which both of them are growing. Artificial infection of *Loranthus* growing on *Bauhinia purpurea* L. also indicates that *Cuscuta* is in a position to rapidly grow on the parasite as well as the main host, and in so doing it does adjust its sap concentration in such a way that a steady diffusion of nutrients may be made possible for its normal growth.

In the light of these and other observations obtained on artificial injection of sugars inside the host tissue it appears that *Cuscuta* can more efficiently absorb nourishment on account of its tendency to rapidly adjust its sap concentration

according to the changes in the nutritional conditions of the host. Parasitism, in its broader sense, therefore, appears to include all those phenomena that characterise plants feeding on either a host growing normally on the soils or a host that is itself growing parasitically. The superposed parasitism of *Cuscuta* over *Loranthus* is an unique example of double parasitism in higher plants. The two types of parasites grow quite harmoniously for a fairly long time, this being made possible on account of the increasing gradient of osmotic and H-ion concentrations so characteristically established from the host-end to that of the parasites. In consequence both *Cuscuta* and *Loranthus* grow well so long as the common host can supply the nourishment to the competing parasites but their gradual withering is apparent soon after that power of the host is lost.

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# THE PREVENTION OF ROTS IN TOMATOES WITH ESPECIAL REFERENCE TO THE MOULD'S ATTACK

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## SUMMARY

1. The investigation has been designed with a view to minimizing the extent of rot in tomatoes which seriously hampers the fruit industry.

2. Two varieties of tomatoes, *Lycopersicum esculentum* (var. Abundance) and *Lycopersicum esculentum* (var. Local) have been worked out with regard to the effect of iodized paper wrap, wood shaving and cork dust on the extent of rotting.

3. Materials are subjected to different temperatures and the way in which they resist the attack of moulds is furnished.

4. An account of the ripening processes with particular reference to colour changes and gaseous exchange in the packed tomatoes is given.

5. It is concluded that iodized wood shaving has a definite advantage over iodized paper wrap in the prevention of rot in tomatoes.

Tomatoes suffer serious wastage in transportation from the growth of fungi and bacteria on their surface. Practical method to reduce this loss is urgently needed.

Two modes of attack are in main suggested; first, to prevent the infection of the fruit and secondly, to store them under conditions detrimental to the growth of the causal organism. The former seems impracticable to apply since the mould spores float freely extensively in the atmosphere and infect the fruits under field conditions, while the latter has been tried with some measure of success.

The dipping of the fruits in disinfecting solutions of borax and potassium permanganate, as well as the addition of ammonia, acetaldehyde and sulphur dioxide to the storage atmosphere have all proved unsatisfactory because of the serious engineering difficulties in the maintenance of the required conditions. Plain wraps have been found to prevent the spread of mycelial growth on fruits but not the growth of the organisms on the fruit surface, and therefore, the especially prepared wraps are preferred. The wraps impregnated with iodine and iodoform have been recently used to prevent the rotting of fruits, while those with copper sulphate<sup>2</sup> for the rotting of pears, and those with mineral oil<sup>1</sup> for the scald in apples. The wraps impregnated with the latter substances neither reduce the incidence of rotting nor retard the growth of the organism but entail wrapping difficulties and high storage cost, as also do the iodized wraps. The iodoform wraps taint the fruits and therefore iodine which does not stain fruits when used in minute doses, was employed for the purpose. The observations herein recorded show the effect of iodized wood shaving, cork dust and paper wrap on the extent of rotting of tomatoes, hoping that the first two materials might prove considerably more economical than the wrapping of the fruits.

#### EXPERIMENTATION

Two varieties of tomatoes, namely, *Lycopersicum esculentum* (var. abundance) and *Lycopersicum esculentum* (var. local) were selected and the fruits which were considered chronologically and physiologically fit at the mature but unripe stage as indicated by Gustafson<sup>1</sup> were employed for detailed investigation. In the main, the fruits were arranged in wood trays with iodized paper wrap, wood shaving and cork dust as the packing materials at 0°, 10°, 20°C and at the laboratory temperature (25°C). The observations were made both with regard to the ripening processes and the percentage of rots in fruits at ten days' intervals. The gaseous exchange was measured by Singh and Mathur's modified Haldane Gas Analysis "Apparatus" while the dissolved carbon-dioxide was determined in accordance with the method described by Willaman and Brown for apple twigs.

The packing materials were prepared by mixing 100gms. of each substance with 1.5 litres of iodine solution, prepared from 13.9 gms. of iodine 11.2 gms. of potassium iodide in 100 c.c. of water and 900 c.c. of rectified spirit. The materials were dried at the ordinary laboratory temperatures before using them for the packing purposes.

## OBSERVATION

*A. The effect of packing material on the rotting percentage:—*The percentages of tomatoes suffering from rots after storage for definite days at different temperatures (Table I) are sufficiently accountable,

Table I

Influence of Temperature and Packing on the rotting percentage in tomato.

Plant material		<i>L. esculentum</i> (var. Abundance)				<i>L. esculentum</i> (var. Local)		days
Temperature in C.	Iodized material	10	20	30	40	20	40	
0°	wood shaving	16	56	71	93	65	99	
	cork dust	17	60	79	94	71	...	
	paper wrap	20	70	86	99	81	...	
10°	wood shaving	8	11	20	50	22	65	
	cork dust	7	18	26	59	26	76	
	paper wrap	8	19	29	69	30	84	
20°	wood shaving	14	20	38	78	35	98	
	cork dust	17	24	44	85	37	...	
	paper wrap	20	30	58	91	41	...	
Laboratory (25° ± 4°)	wood shaving	25	50	69	89	75	...	
	cork dust	27	33	73	96	73	...	
	paper wrap	29	65	82	...	85	...	

leading one to believe that the fruits stored in iodized wood shaving store better than the iodized paper wrapped fruits or the fruits kept in iodized cork dust. The tough nature of the shaving might reduce the rotting

percentage since it gives an advantage over others by way of diffusing free iodine comparatively slowly. The percentage of loss increases in general with an increase in the storage duration. A decrease in temperature shows a concomitant diminution in the incidence of the mould and the appearance of the physiological diseases (Table II), and as such it decreases the percentage of rotting with low temperatures. This is, however, not true for very low temperatures where low temperature injury<sup>7</sup> intervenes the metabolic rates and freezes the fruit tissues to physiological rot. At 10° C. the percentage of rotting is the least and the fruits store best suggesting that in the vicinity of 10° C. the activity of the parasite is considerably reduced and the physiological decay is diminished. The variety *Abundance* suffers more from rots than the local variety and also has a short storage life. The incidence of the mould on appearance shows that, in general, it starts from the calyx end or from the blushed portions of the fruit. A luxurious growth of *Rhizopus* is often noticed near the fruiting twig at the latter stages of the storage life.

Having observed the incidence of moulds, the fruits were examined with regard to their gaseous exchange assuming that the latter is associated with the protoplasmic activity. It is hoped that the study of the gaseous exchange shall visualize the processes of ripening and the concurrent incidence of the storage diseases under the experimental conditions, and shall ultimately help to develop a method for the prevention of rots in tomatoes.

*B. The effect of packing materials on the processes ripening :—*The effect of packing material was more pronounced on colour changes during storage than the recorded figures could suggest. The fruits, in general, changed their colours as ripening proceeded from green to orange yellow, orange-red and red in regular succession. During this process some fruits gave off ethylene or a substance having identical properties of ripening as shown by Regeimbal and others<sup>8</sup> on bananas, at a certain stage of ripeness, which set off all the others with it. Working on apples in the temperate region a similar observation was made by Kidd<sup>1</sup>. The smell and taste of the individual healthy fruits was not affected but the smell of fruit which usually pervades the storage atmosphere was destroyed.

The changes in the colour of the fruits were concomitant with the changes in the economy of the gaseous exchange. The rate of respiration of the green tomatoes, in general, increases as the colour changes from green to red and then decreases till the fruits become red ripe followed by a burst of activity before death.

Table II

Gaseous exchange in tomatoes during storage

Temperature in C.	Colour of the fruit	CO <sub>2</sub> liberated ml. / kilo / hr.		Dissolved CO <sub>2</sub> ml. per kilo		R.Q.
		<i>var.</i> Abun- dance	<i>var.</i> local	<i>var.</i> Abun- dance	<i>var.</i> local	
Iodized wood shaving						
0°	Green (healthy)	6.3	4.4	30.9	26.6	...
	Orange-green (dead)	4.5	3.3	43.1	39.1	...
10°	Green	10.0	7.3	28.6	24.2	...
	Orange-yellow	13.3	9.8	34.9	30.1	...
	Red	14.0	10.0	...	...	...
	Red ripe	8.4	5.0	38.8	34.6	...
	Red soft	4.4	2.3	40.0	36.0	...
20°	Green	15.5	12.2	23.3	15.9	1.11
	Orange-yellow	18.8	14.9	30.6	25.9	1.01
	Red	19.1	15.2	...	...	...
	Red ripe	14.6	11.5	33.2	29.1	1.27
	Red soft	8.9	5.4	38.1	32.6	1.29
Laboratory (25° ± 4°)	Green	17.7	13.6	19.7	15.6	...
	Orange-yellow	20.7	18.0	28.6	24.1	...
	Red	21.3	18.8	...	...	...
	Red ripe	16.8	14.4	30.3	25.9	...
	Red soft	10.6	9.9	36.5	32.3	...
Iodized paper wrap						
20°	Green	13.3	10.3	30.9	26.6	...
	Orange-yellow	16.5	12.0	38.0	33.0	...
	Red	17.0	12.3	...	...	...
	Red ripe	12.0	9.3	41.1	37.9	...
	Red soft	7.2	4.0	46.3	40.0	...

The respiratory quotient figures start, in general, with a value higher than unity, come down to unity and again rise till death. The values are sensibly similar in the iodized wood shaving and cork dust at different temperatures and therefore it is not possible to give the figures for them separately. The dissolved



CO<sub>2</sub> continues to increase during storage. The values of CO<sub>2</sub> out-put obtained for the iodized paper wraps are lower than the other two treatments while the wrapping increases the concentration of the dissolved CO<sub>2</sub>. The fruits, thus, suffer from physiological diseases comparatively severely. The incidence of mould is more at the red-stage than in the green stage. It may be due to a very rapid metabolic rate of the fruit tissues which increases the virulence of the storage diseases.

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# AN ANOMALY IN THE ELASTIC BEHAVIOUR OF INDIA RUBBER

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## SUMMARY

The Young's modulus and rigidity for a sample of India Rubber are determined. Young's modulus is measured both statically and dynamically. It is found that the dynamical value is about twice the statical value for all loads. The cause of this anomaly is observed but no satisfactory explanation has been found.

## INTRODUCTION

The elastic properties of India Rubber have been studied by a number of investigators<sup>1,2</sup>. The elastic moduli for India Rubber are much smaller than for metals and the periodic times in any vibration experiment are sufficiently large to be studied accurately. Therefore the study of elastic behaviour of India Rubber offers a good field of investigation in elucidating some interesting problems which it would be very difficult, if not impossible, to study in the case of metals. The object of the present paper is to study one such problem. In the study of the rigidity and Young's modulus of India Rubber, Deodhar and Kothari<sup>3</sup> observed a fairly large discrepancy between the observed values of rigidity (determined by Maxwell's method) and the value calculated from Young's modulus (determined statically) and Poisson's ratio, in accordance with the well known formula  $\eta = \frac{E}{2(1+\sigma)}$ .

It was found that the observed rigidity was larger by a factor of about 2 than the calculated value for all loads up to the breaking stress. This discrepancy between the calculated and observed values of rigidity is (it appears)

no special property of India Rubber alone but is characteristic of the metals as well, though to a much less extent. The following table brings out this fact clearly.

Table I

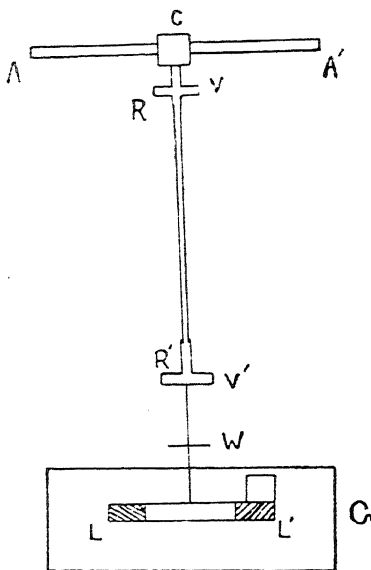
Substance	$\eta$ oscillation method	$\eta' = \frac{E}{2(1+\sigma)}$	$\frac{\eta \text{ (observed)}}{\eta' \text{ (calculated)}}$
Aluminium	$2.67 \times 10^{11}$	$2.63 \times 10^{11}$	1.02
Zinc	$3.8 \times 10^{11}$	$3.59 \times 10^{11}$	1.06
Copper	$4.55 \times 10^{11}$	$4.55 \times 10^{11}$	1.00
German Silver	$4.50 \times 10^{11}$	$4.25 \times 10^{11}$	1.05
Gold	$2.77 \times 10^{11}$	$2.80 \times 10^{11}$	.99
Platinum	$6.10 \times 10^{11}$	$6.04 \times 10^{11}$	1.01
Bronze	$3.43 \times 10^{11}$	$2.97 \times 10^{11}$	1.15
Quartz	$3.0 \times 10^{11}$	$2.61 \times 10^{11}$	1.15

From the above tables it is evident that with the exception of gold,  $\eta$  is in all other cases never less and generally greater than  $\eta'$ . India Rubber in which this discrepancy between  $\eta$  and  $\eta'$  occurs to such a great extent, offers the best material for elucidating the nature of this discrepancy. The present study was undertaken with this object in view.

After a careful study of the subject it appeared that the cause of this discrepancy lies in the fact that while  $\eta$  is determined by dynamical methods,  $E$  is measured by statical methods. This can be tested by determining  $E$  also dynamically. The experiments, given below show that  $\eta$  calculated from the dynamical values of  $E$ , agree reasonably well with the observed values for all loads. The apparatus designed for carrying out the experiments is as follows :—

## PROCEDURE

The apparatus is essentially a Maxwell needle and is very simple in construction. An iron bar  $AA'$  is fixed firmly at its ends in a wall and in the middle of the bar, there is a clamp  $C$ , in which is fixed a vice  $V$  which holds one end of the India-Rubber band  $RR'$  firmly. The other end of the band is clamped in another vice  $V'$  to the lower end of which is screwed an iron rod carrying a weight pan  $W$  at its end, to hold a number of weights. To the lower end of the pan is soldered a Maxwell needle  $LL'$  which is enclosed in a box with glass sides, to avoid air draughts. In the centre of the needle is fixed a plane mirror which is used for measuring the period of vibration with the help of a lamp and a scale. The length of the rubber band was measured by a kathetometer reading up to '01 cm. while the diameter was measured with a screw gauge. A number of equidistant marks were made on the rubber band with ink and the readings of the diameter of the band were taken at these places. Two other marks were made on the band, one near the top and the other near the bottom and the distance between them was measured for determining the value of Young's modulus.



To start with the experiment, a load of one K. gm. was put in the weight pan and the length of the rod  $RR'$  determined. Then the length of the rod between the two marks was measured. After that five readings of the diameter were taken on alternate marks and the period of vibration for vibrations in the horizontal plane determined with the solid tubes outside and the empty tubes inside the tube. Then the rubber rod was made to vibrate up and down and its period of vibration determined. For this a very simple device was used. A piece of card board was fixed to the centre of the needle and its shadow which was produced by a slit, illuminated by a lamp and placed behind the needle, was cast on a ground glass plate. In that way the period was determined very accurately. After this the period of vibration in the horizontal plane with the solid tubes inside and the empty tubes outside, was determined. Then a weight of one K. gm. was added to the weight pan

and after some time (about 30 minutes) the period of vibration of the up and down vibrations was determined. The length between the two marks was again determined and five readings of the diameter taken on the remaining marks. This method eliminated to some extent the changes in length and diameter taking place during the time that elapses in taking the necessary observations. The load was increased by steps of one K. gm. each till 9.5 K. gm. was reached when it was found that the period of vibration in the vertical plane became very small and so further observations had to be stopped. The observations were recorded according to the following plan.

*Observations for 4.5 K. gm. Load.*

Temperature	...	...	...	...	30°C
Length of the band RR'	...	...	...	$L_1 =$	40.18 cm.
Distance between the two marks	...	...	...	$L_2 =$	23.54 cm.
Diameter...	...	...	...	$d =$	5.34 m.m.
Period of oscillation with solid tubes outside	...	...	...	$T_1 =$	41.07 Secs.
"    "    "    in vertical plane	...	...	...	$t_1 =$	.752 Sec.
"    "    "    with solid tubes inside.....	...	...	...	$T_2 =$	30.0 Secs.
"    "    "    in vertical plane with constant mass	...	...	...	$t_2 =$	.692 Sec.
Diameter	...	...	...	$d =$	5.09 m.m.
Length between the marks	...	...	...	$L_2 =$	25.85 cm.
Increase in length $= l = L_2 - L_1$	...	...	...	$=$	2.31 cm.
Then Young's modulus under statical conditions was determined by the ordinary formula.					

$$E = \frac{F}{a} \times \frac{L}{l} = 468.9 \times 10^5 \text{ dynes/cm}^2.$$

where  $a = \pi r^2$ ,

$r$  being the radius of the band.

The rigidity  $\eta$  was determined by the following formula

$$\eta = \frac{2\pi x^2 L_1 (m_1 - m_2)}{r^4 (T_1^2 - T_2^2)} = 354.8 \times 10^5 \text{ dynes/cm}^2.$$

where

$x$  = length of Maxwell needle.

$m_1$  = mass of the solid tubes.

$m_2$  =       "      empty tubes.

The value of dynamical Young's modulus was calculated as follows:—

$$E = \frac{4 \pi L (M_1 - M_2)}{r^2 (t_1^2 - t_2^2)} = 867.2 \times 10^5 \text{ dynes/cm}^2.$$

where  $M_1$  and  $M_2$  are the masses for periods  $t_1$  and  $t_2$  respectively.

For each load observations were taken as described above and the results calculated. The results for various stresses are given below. Here  $\eta$  is the rigidity determined experimentally,  $\eta'$  is the value of rigidity calculated from the static value of Young's modulus by applying the formula  $\eta' = \frac{E}{2(1+\sigma)}$  when  $\sigma$  is Poisson's ratio.  $\eta''$  is the value of rigidity calculated from the dynamical value of Young's modulus by the same formula.

### RESULTS

Table II

Stress dynes/cm <sup>2</sup>	E (Statical) dynes/cm <sup>2</sup>	$\eta$ (observed)	E' (dynamical) dynes/cm <sup>2</sup>	$\sigma$ Poissons ratio	$\eta'$ $\frac{E}{2(1+\sigma)}$	$\eta''$ $\frac{E'}{2(1+\sigma)}$	$\frac{\eta}{\eta'}$	$\frac{\eta}{\eta''}$
$1.47 \times 10^6$	$206.8 \times 10^5$	$133 \times 10^5$	$277.9 \times 10^5$	.34	$77.2 \times 10^5$	$103.4 \times 10^5$	1.72	1.29
$2.45 \times 10^6$	$277.3 \times 10^5$	$173.8 \times 10^5$	$526.0 \times 10^5$	.40	$99.0 \times 10^5$	$187.9 \times 10^5$	1.75	.90
$3.43 \times 10^6$	$390.4 \times 10^5$	$245.5 \times 10^5$	$622.0 \times 10^5$	.50	$125.9 \times 10^5$	$207.3 \times 10^5$	1.95	1.18
$4.41 \times 10^6$	$468.9 \times 10^5$	$354.8 \times 10^5$	$867.2 \times 10^5$	.47	$159.5 \times 10^5$	$294.9 \times 10^5$	2.22	1.18
$5.39 \times 10^6$	$568.0 \times 10^5$	$428.3 \times 10^5$	$1199.4 \times 10^5$	.46	$195.0 \times 10^5$	$410.6 \times 10^5$	2.19	1.04
$6.37 \times 10^6$	$664.5 \times 10^5$	$569.0 \times 10^5$	$1708.7 \times 10^5$	.48	$224.9 \times 10^5$	$577.7 \times 10^5$	2.08	.98
$8.33 \times 10^6$	$1096 \times 10^5$	$714.5 \times 10^5$	$2014.3 \times 10^5$	.53	$358.1 \times 10^5$	$658.2 \times 10^5$	1.99	1.08
$9.31 \times 10^6$	$1406 \times 10^5$	$1007 \times 10^5$	$2404 \times 10^5$	.51	$465.5 \times 10^5$	$796.6 \times 10^5$	2.14	1.27

It is clear from the above tables that the dynamic value of Young's modulus is much larger than the static value and that the values of rigidity calculated from the dynamical values of E agree very closely with those

determined experimentally under dynamic conditions. Hence the discrepancy in the case of India Rubber may be easily explained as due to the fact that  $E$  and  $\eta$  are measured under different conditions.

#### DISCUSSION

The significant fact that emerges out from our observations is that the dynamical  $E$  is much larger than the statical value of  $E$ . A theoretical explanation which immediately suggested itself was that the dynamical  $E$  corresponds to the adiabatic modulus and the statical  $E$  to the isothermal one. As is well known from the usual thermodynamical arguments,  $E_i$  and  $E_A$  are connected by the relation,<sup>1</sup>

$$\frac{1}{E_i} - \frac{1}{E_A} = \frac{\alpha^2 T}{J S \rho}$$

where  $T$  is the absolute temperature,  $\rho$ , the density, and  $\alpha$  the coefficient of linear expansion and  $S$  the specific heat.

For the sample of rubber used,  $S$  was found to be .42, and  $\rho$  to be .92. The value of  $\alpha$  was experimentally determined, both at low and high temperatures at a load of 3.5 K. gm. As a result of a number of experiments it was found that the band contracted up to 75°C, but beyond that temperature it continued to increase in length. In the former case the value of  $\alpha$  was found to be -.00054 while in the latter case it was found to be +.0033. On substituting the values of  $T, S$ , and  $\rho$  in the above formula, the value of  $\frac{E_A}{E_i}$  was found to be 1.0004, if the value of  $\alpha$  be taken as -.00054, but the ratio is found to be 1.04 if the value of  $\alpha$  be taken as .0033. In order that the value of the ratio may agree even roughly with the observed value  $\alpha$  ought to be about 1/60. Hence this anomaly in the elastic behaviour of India Rubber, in that the statical and dynamical  $E$  differ by a factor of about 2, cannot be accounted on purely thermodynamical grounds but it probably has something to do with the constitution of India Rubber and for this purpose a fresh study of the subject will be undertaken very shortly. However, the present experiment clears the discrepancy noted by Deodhar and Kothari with regard to the observed and calculated rigidity. This discrepancy arose because the rigidity was measured by a dynamical method, whereas in the relation

$$E = \frac{E}{2(1+\sigma)}$$

the value of  $E$  was determined statically. We have seen that if we use for  $E$  the value determined dynamically, then the observed rigidity

agrees within the limits of the experimental error with the value calculated from the above relation.

Thanks are due to Dr. D. S. Kothari for interest in this work.

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# CURVED ASYMPTOTIC LINES OF RULED SURFACES

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1. The object of this paper is to obtain some properties of the curved asymptotic lines of ruled surfaces.

Let the equations of a ruled surface be  $x = p + lu$ ,  $y = q + mu$ ,  $z = r + nu$ , where  $p, q, r, l, m, n$  are functions of  $v$ , the arc of the base curve. The differential equation of the curved asymptotic lines of the ruled surface is

$$\frac{du}{dv} + \frac{\lambda + \mu u + \nu u^2}{2\delta} = 0, \text{ where}$$

$$\lambda = \Sigma l (q' r'' - r' q''), \mu = \Sigma l \{ (m' r'' - m'' r') + (q' n'' - n' q'') \},$$

$$\nu = \Sigma l (m' n'' - n' m''), \delta = \Sigma l (q' n' - r' m'),$$

accents denoting differentiation with regard to  $v$ .

2. We shall first consider the curved asymptotic lines of those ruled surfaces whose generators are parallel to a fixed plane.

Let the fixed plane be taken as the plane of  $xy$ , and let a curved asymptotic line be taken as the directrix. The equations of the ruled surface can be written as  $x = p + m_3 u$ ,  $y = q - l_3 u$ ,  $z = r$ , where  $l_3, m_3, n_3$  are the direction cosines of the binormal of the directrix curve. Also let  $l_1, m_1, n_1; l_2, m_2, n_2$  be the direction cosines of the tangent and the principal normal of the directrix curve.

Now since  $\lambda = 0$  and  $\nu = 0$ , the equation of the curved asymptotic lines reduces to

$$\frac{du}{dv} + \frac{\mu}{2\delta} u = 0 \quad \dots \quad (1)$$

But using Frenet's formulae

$$\begin{aligned}\delta &= \Sigma l (q' n' - r' m') = l (q' n' - r' m') + m (r' l' - p' n') + n (p' m' - q' l') \\ &= m_3 \left( -n_1 \cdot \frac{l_2}{\sigma} \right) + (-l_3) \left( n_1 \cdot -\frac{m_2}{\sigma} \right) = \frac{1}{\sigma} \cdot n_1 (l_3 m_2 - l_2 m_3) = \frac{n_1^2}{\sigma} \dots \quad (2)\end{aligned}$$

$$u = l \{ (m' r'' - m'' r') + (q' n'' - n' q'') \} + m \{ (n' p'' - n'' p') + (r' l'' - l' r'') \} \\ + n \{ (l' q'' - l'' q') + (p' m'' - m' p'') \}$$

$$= m_3 \left[ \frac{l_2}{\sigma} \cdot \frac{n_2}{\rho} - n_1 \left\{ \frac{1}{\sigma} \left( \frac{l_3}{\sigma} - \frac{l_1}{\rho} \right) + l_2 \frac{d}{dv} \left( \frac{1}{\sigma} \right) \right\} \right] \\ + (-l_3) \left[ n_1 \frac{d}{dv} \left( -\frac{m_2}{\sigma} \right) - \left( -\frac{m_2}{\sigma} \right) \left( \frac{n_2}{\rho} \right) \right]$$

$$= (-m_3 n_1 l_2 + l_3 n_1 m_2) \frac{d}{dv} \left( \frac{1}{\sigma} \right) + \frac{1}{\rho \sigma} (m_3 l_2 n_2 + m_3 n_1 l_1 - m_1 n_1 l_3$$

$$- l_3 m_2 n_2) + \frac{1}{\sigma^2} (-m_3 l_3 n_1 + m_3 l_3 n_1)$$

$$= (-m_3 n_1 l_2 + l_3 n_1 m_2) \cdot \frac{d}{dv} \left( \frac{1}{\sigma} \right) = n_1^2 \frac{d}{dv} \left( \frac{1}{\sigma} \right), \text{ since}$$

$$\text{coefficient of } \frac{1}{\rho \sigma} = m_3 (l_2 n_2 + n_1 l_1) - l_3 (m_1 n_1 + m_2 n_2) = m_3 (-n_3 l_3)$$

$$- l_3 (-m_3 n_3) = 0.$$

$$\therefore (1) \text{ becomes } \frac{du}{dv} + n \cdot \frac{\sigma}{2} \frac{d}{dv} \left( \frac{1}{\sigma} \right) = 0, \text{ whose solution is}$$

$$u = c \sqrt{\sigma} \dots \dots \dots (3)$$

(*Nouvelle Annales de Mathematiques*, 1924)

where  $c$  is an arbitrary constant and  $\sigma$  is the radius of torsion of the directrix curve.

If  $t$  be the distance between the points where two asymptotics cut a variable generator, and if  $\theta_1, \theta_2$  be the inclinations of the binormals to the  $z$ -axis, then since  $t^2 = u^2 (l_3^2 + m_3^2) = u^2 \sin^2 \theta$ , we get

$$t = c_1 \sqrt{\sigma_1} \cdot \sin \theta_1, \text{ and}$$

$$t = c_2 \sqrt{\sigma_2} \cdot \sin \theta_2.$$

Hence  $\frac{\sin \theta_1 \cdot \sqrt{\sigma_1}}{\sin \theta_2 \cdot \sqrt{\sigma_2}} = \text{constant for any two curved asymptotics of a ruled surface}$

whose generators are parallel to a fixed plane.

The torsion of the directrix curve being known, the torsions of all the other curved asymptotics can be determined as follows:—

The radius of torsion of an asymptotic line by Beltrami-Eunepers' formula<sup>5</sup> is  $\pm \sqrt{\frac{-(EG-F^2)}{LN-M^2}}$ , where E, F, G, and L, M, N are the fundamental magni-

tudes of the first and second order respectively, or  $\pm \frac{\sqrt{EG-F^2}}{M}$  since  $L=0$ , or

$$\pm \frac{EG-F^2}{MV}, \text{ since } V = \sqrt{EG-F^2}, \text{ or } \pm \frac{EG-F^2}{\delta}.$$

$$\begin{aligned} \text{But } E &= m_3^2 + l_3^2 = 1 - n_3^2; G = 1 + \frac{u^2}{\sigma^2}(m_3^2 + l_3^2) + \frac{2u}{\sigma}(l_1m_2 - l_2m_1) \\ &= 1 + \frac{u^2}{\sigma^2}(1 - n_3^2) + \frac{2un_3}{\sigma}; F = n_2 \left( \frac{-un_3}{\sigma} - 1 \right). \\ \therefore EG - F^2 &= n_1^2 + u \left( \frac{2n_3}{\sigma} - \frac{2n_3^3}{\sigma} - 2n_2^2 \cdot \frac{n_3}{\sigma} \right) + u^2 \left\{ -\frac{1}{\sigma^2}(1 - n_3^2)n_2^2 - n_2^2 \cdot \frac{n_3^2}{\sigma^2} \right. \\ &\quad \left. + \frac{1}{\sigma^2}(1 - n_3^2) \right\} = n_1^2 \left( 1 + \frac{2n_3}{\sigma}u + \frac{n_1^2}{\sigma^2}u^2 \right). \end{aligned}$$

Hence the radius of torsion of any curved asymptotic line  $= \frac{EG-F^2}{\delta} = \sigma + 2n_3u$

$$+ \frac{u^2}{\sigma} = \sigma + 2n_3c\sqrt{\sigma} + c^2 \text{ from (3),} \quad \dots \quad (4)$$

(Nouvelle Annales de Mathematiques, 1924)

where c is the arbitrary constant corresponding to that asymptotic line.

If two curved asymptotic lines have the same torsion then we have  $\sigma = \sigma + 2n_3c\sqrt{\sigma} + c^2$ , which is a quadratic in c and hence shows that there cannot be more than two curved asymptotic lines on the ruled surface whose generators are parallel to a fixed plane which have the same torsion at the points where a generator cuts them.

3. Some properties of the curved asymptotic lines of any ruled surface can be obtained from Beltrami-Enneper's formula, and the formula for the total curvature viz.

$$K = - \frac{\beta^2}{[(u-a)^2 + \beta^2]^2}$$

where  $\beta$  is the parameter of distribution and  $u-a$  is the distance from the central point.

Along a generator  $K$  takes each value twice at most and hence at most two of the curved asymptotics can have their torsions equal to one another at the points where a generator cuts them.

From  $K = - \frac{\beta^2}{[(u-a)^2 + \beta^2]^2}$ , it follows that if two curved asymptotic lines have the same torsion where they are met by a generator, so that  $K$  has the same value at these points, then the points are equidistant from the central point, since  $K$  takes each value twice only.

Hence

The line of striction of the ruled surface which has two curved asymptotic lines with the same torsion at the points where they are cut by a generator is the locus of the middle points of the lines joining the corresponding points.

Also, since the middle point of the segment intercepted on a generator by any two orthogonal trajectories describes another orthogonal trajectory, it follows that :

If two curved asymptotic lines with the same torsion on a ruled surface be orthogonal trajectories of the generators, then the line of striction of the surface is also an orthogonal trajectory of the generators.

4. Some properties of the curved asymptotic lines can also be deduced by considering the ruled surfaces formed by joining the corresponding points on two curves  $C_1$  and  $C_2$ , the points on which are in one to one correspondence in such a way that the lines joining the corresponding points of  $C_1$  and  $C_2$  lie simultaneously in the osculating planes of  $C_1$  and  $C_2$  at the corresponding points. Such curves  $C_1$  and  $C_2$  have been called the asymptotic transformations of each other by Bianchi.<sup>2</sup>

Let  $P_1(x_1, y_1, z_1)$  be a point on  $C_1$  and  $P_2(x_2, y_2, z_2)$  its corresponding point on  $C_2$ ; also let  $t$  be the distance between  $P_1$  and  $P_2$ ,  $\theta$  the angle which  $P_1 P_2$  makes with the tangent to  $C_1$  at  $P_1$ , and  $\sigma$  the angle between the osculating planes of the two curves at  $P_1$  and  $P_2$ . Then  $t, \theta, \sigma$  are functions of an independent variable  $u$  which is the arc of  $C_1$  measured from a fixed point on it. Denoting the direction cosines of the tangent, principal normal and binormal to  $C_1$  at  $P_1$  and to  $C_2$  at  $P_2$ , by  $l_1, m_1, n_1; l_2, m_2, n_2; l_3, m_3, n_3$  and the corresponding letters with accents we have  $x_2 = x_1 + t(l_1 \cos \theta + l_2 \sin \theta)$ , with similar expressions for  $y_2, z_2$ ; and  $l_3' = l_1 \sin \sigma \sin \theta - l_2 \sin \sigma \cos \theta + l_3 \cos \sigma$ , with similar expressions for  $m_3'$  and  $n_3'$ .<sup>3,7,8</sup>

Now it has been proved by Tortorici<sup>8</sup> that if one curve  $C_1$  is given, in order that another curve  $C_2$  be its asymptotic transformation,  $t, \sigma$  and  $\theta$  must satisfy the two relations

$$\Sigma l_3' \frac{dx_2}{du} = \frac{d\theta}{du} + \frac{1}{\rho_1} - \sin \theta \cdot \left( \frac{1}{t} - \frac{\cot \sigma}{\sigma_1} \right) = 0 \quad \dots \quad (5)$$

$$\text{and } \Sigma \frac{dl_3'}{du} \cdot \frac{dx_2}{du} = \cos \theta \left( \frac{-t^2}{\sigma_1^2} + \sin^2 \sigma \right) + \frac{t^2}{\sigma_1} \cdot \frac{d\sigma}{du} + \sin^2 \sigma \cdot \frac{dt}{du} = 0 \quad \dots \quad (6)$$

We shall first give an independent proof of the converse viz. that (5) and (6) are sufficient conditions that a line through  $P_2$  perpendicular to  $P_1 P_2$  and making an angle  $\sigma$  with  $(l_3, m_3, n_3)$  should be the binormal of the locus of  $P_2$ , so that the locus of  $P_2$  having its binormal perpendicular to  $P_1 P_2$  is an asymptotic on the ruled surface described by  $P_1 P_2$  :—

Let  $(l_3', m_3', n_3')$  be the normal at  $P_2$  to the ruled surface generated by  $P_1 P_2$  and let it make an angle  $\sigma$  with  $(l_3, m_3, n_3)$ ; then  $l_3' = l_1 \sin \sigma \sin \theta - l_2 \sin \sigma \cos \theta + l_3 \cos \sigma$ , .... Since (5) is satisfied we have  $\Sigma l_3' l_1' = 0$ , i. e.,  $(l_3', m_3', n_3')$  is perpendicular to  $(l_1', m_1', n_1')$ . Since (6) is satisfied we have  $\Sigma \frac{dl_3'}{du} \cdot l_1' = 0$ .

$$\text{Differentiating } \Sigma l_3' l_1' = 0 \text{ we get } \Sigma \frac{dl_3'}{du} l_1' + \Sigma l_3' \frac{dl_1'}{du} = 0$$

$\therefore \Sigma l_3' l_2' = 0$ ,  $\therefore (l_3', m_3', n_3')$  is perpendicular to  $(l_2', m_2', n_2')$  also. Hence  $(l_3', m_3', n_3')$  is the binormal of  $C_2$  at  $P_2$ . Hence  $C_2$  is an asymptotic on the ruled surface generated by  $P_1 P_2$ . (N. B.  $u$  has been taken as the arc of  $C_2$  in this proof).

We shall now deduce one property of the curved asymptotic lines.

If  $\sigma_1$  and  $\sigma_2$  are the radii of torsion of the two curves  $C_1$  and  $C_2$  at  $P_1$  and  $P_2$ , then  $\frac{1}{\sigma_1} \cdot \frac{1}{\sigma_2} = \frac{\sin^2 \sigma}{t^2}$  ... (7)

$\therefore$  if  $\frac{1}{\sigma_1} = \frac{1}{\sigma_2} = \text{constant} = -1$ , say, for convenience, then (5), (6) and (7) are equivalent to

$$\sigma = \text{constant} = a, \text{ say : } t = \sin a.$$

$$\frac{d\theta}{du} + \frac{1}{\rho_1} = \sin \theta. (\operatorname{cosec} a + \cot a).$$

If  $a$  is chosen arbitrarily,  $\theta$  depends on  $a$  and on a constant of integration. There are consequently  $\infty^2$  ruled surfaces through the given curve  $C_1$ , each of which contains a second asymptotic having also its torsion equal to  $-1$ . On any one of these ruled surfaces, there is only one such second asymptotic because  $t$  is fixed by  $t = \sin a$ . Hence we get the result

'A ruled surface cannot have more than two curved asymptotic lines having the same torsion at the points where they are cut by a generator.'

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## AZINE COMPOUNDS DERIVED FROM DI-AMIDO-BENZIDINE

By L. D. TIWARI AND S. DUTT.

### SUMMARY

Interesting dyestuffs have been prepared by the condensation of 3:4:3':4'-tetraamidodiphenyl for the first time with ortho-diketones like *p*-naphthoquinone, acenaphthoquinone, phenanthraquinone, isatin, benzil etc. All these condensation products develop intense colorations with concentrated sulphuric acid, which can be explained in accordance with a theory of colour on the basis of molecular strain advanced by one of the present authors. All these condensation products dye wool in different shades of yellow from an acid bath.

Azines and azonium compounds have been prepared by condensation of orthodiamines with orthodiketones. A large number of such condensation products which are of technical importance on account of their tinctorial property have already been described in the literature.<sup>6,9,13,14,15</sup> Thus it was found that the more complex the orthodiamine the darker the colouring matter obtained by its condensation with the orthodiketones. With this point in view, azine compounds were prepared by the condensation of diamidobenzidine with the orthodiketones.

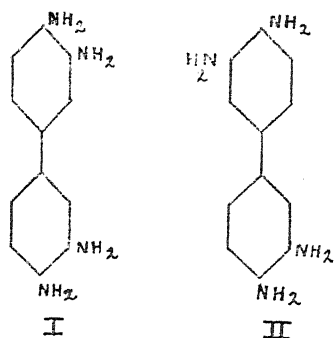
The complexity of the orthodiamine here has been enhanced in more than one way. Firstly, there are four amine groups in two benzene nuclei in ortho positions hence one more molecule of the diketone is likely to enter in the reaction. Secondly, the situation of the two amino groups in the two benzene nuclei would affect the symmetry of the molecule which in its turn shall have considerable effect on the colour of the azines formed. Colour varies directly as the strain, according to Dutt<sup>7,8</sup> e. g. Fluorescein (yellow)  $\lambda$  4940, Dibromofluorescein (orange)  $\lambda$  5050.

The same result can be inferred from the fact that orthoquinonoid compounds are more coloured than paraquinonoid that is the former are unsymmetrical and more strained and the latter are symmetrical and less strained, e. g. parabenzeneazophenol (yellow)  $\lambda$  4330; orthobenzeneazophenol (orange red)  $\lambda$  5010.

Tetraamidodiphenyl which has been used in these investigations was prepared by nitrating diacetylbenzidine<sup>16</sup> and then reducing the dinitro product

obtained<sup>3</sup> was condensed with various diketones (ortho) substituted aldehydes and catechol.

Examination of the benzidine formula suggested the possibility of the existence of two tetra-amido-diphenyls as shown below



and the expectations were that (I) would give more coloured azines than (II) (on the basis of strain theory as already stated above).

On consulting the existing literature on these tetraamidodiphenyls, it was found that there have been considerable controversy as regards their existence as isomeric compounds. However, the conclusions arrived at are given below.

Brunner and Witt<sup>4</sup> prepared the azoimide and quinoxaline (with benzil) of the tetraamidodiphenyl and agreed with the Strakosch's formula of the compound which is II above.

Bandrowski<sup>1</sup> prepared tetraamidodiphenyl in a different way, and it was considered to be identical with Strakosch's compound. Cain, Coulthard, and Micklethwart<sup>5</sup> assigned formula (I) above to Bandrowski's compound. Cain and his coworkers showed that benzil condensation product of (I) and (II) were the same. Further the existence of this kind of isomerism among diphenyl compounds have been shown by the work of Kenner and his collaborators<sup>10</sup>.

Brady and McHugh<sup>2</sup> proved the distinct separate existence of the hydrochlorides and stated that they are not interconvertible in any case excepting one, and that being the case of formation of quinoxaline with benzil. On the basis of the experiments of Dennett and Turner<sup>17</sup> Le Fevre and Turner's<sup>12</sup> work has established beyond doubt that Bandrowski's



compound is 4-2:3'-4' and not 4-3:3'-4' as previously supposed. Lastly, Le Fevre, Moir and Turner<sup>11</sup> have shown that the presence of the similar benzil condensation product is due to the presence of 3:3' compound which is present there as impurity and is inseparable under the conditions of preparation of Bandrowski's compound.

Now it becomes quite clear that the compound used in these experiments is 3:3' and the compound 3:5' still awaits the preparation.

3:3' diamidobenzidine was condensed with a number of reactive compounds and the condensation products thus obtained range in colour from dark brown to jet black and show remarkable resistance towards oxidation, chemical decomposition or combustion. In fact, during the process of their analysis for the estimation of nitrogen they required ten to fifteen hours for complete evolution of nitrogen when mixed with a large proportion of copper oxide and heated to bright red heat. Such resistance towards decomposition is possessed by very few classes of organic compounds and subsequently, they may be regarded as some of the most stable organic substances.

The following orthodiketones were condensed with 3-3' tetra-amido-di phenyl and the corresponding azines obtained :— $\beta$  naphthaquinone, acenaphthaquinone, phenanthraquinone, isatin,  $\alpha$ - $\alpha'$  di-cyano-di-keto-di-benzyl, meta-nitro isatin, parabanic acid, alloxan, diacetyl, glyoxal, orthonitrobenzaldehyde, paratolylaldehyde, orthochlorobenzaldehyde and catechol.

The condensations took place easily in presence of glacial acetic acid at the boiling temperature of the solvent except in case of alloxan where condensation was carried out in aqueous medium with sodium acetate. The products were generally precipitated out of the solution in a crystalline condition as soon as the condensation was complete. In a few cases, the condensation product had to be isolated by the concentration of the mother liquor.

The behaviour of these substances towards concentrated sulphuric acid is very characteristic, whereby an intense colour is developed generally in the case of these compounds where the nitrogen atoms are directly linked to the fused aromatic nuclei; the colour is less pronounced in cases where the nitrogen is linked through aliphatic residues or directly to aliphatic radicles as compared to the former. This is quite in agreement with the theory of colour on the basis of molecular strain put forward by

Dutt<sup>7,8</sup>. Nearly all these quinoxaline compounds are dyes without auxomohromes like flavinduline. The colour in these compounds is due to quinoxaline group which is enhanced due to the complex radicle attached to it. The intensification of the colour on addition of concentrated sulphuric acid is due to the nitrogen of the azine group becoming pentavalent. According to Hantzsch's conception of the configuration of nitrogen atom, the valency directions of the tervalent nitrogen are considerably distorted in course of formation of the pentavalent state. Hence the pentavalent nitrogen, apart from the additional load, is more strained than the tervalent one and this is greatly manifested in the greatly increased absorption of pentavalent nitrogen compound e.g. 2-7diamidophenazine  $\lambda$  5150, Phenosafranine  $\lambda$  5250 Phenanthraphenazine  $\lambda$  4300, Flavinduline 4520.

So the strong colour developed when these compounds are dissolved in concentrated sulphuric acid may be regarded to be due to the transformation of a tervalent nitrogen to the pentavalent state by addition of sulphuric acid radicle just as in the case of azo and azine compounds, e.g.

Benazineazophenol (in alcohol)  $\lambda$  4330 (in conc. sulph. acid)  $\lambda$  4560.

Phenazine (in alcohol)  $\lambda$  4290 (in conc. sulphuric acid)  $\lambda$  5630.

But these pentavalent conditions of nitrogen being unstable in dilute solutions, they pass back into the original state by diluting the sulphuric acid solutions. This is exactly the case with the azine compounds that have been synthesized. These substances have no melting point but when strongly heated, they sublime with partial decomposition. From the sulphuric acid solution, the dyestuff can be reprecipitated by the addition of water in a very finely divided form in which state they get readily absorbed by woolen fabric whereby the latter is dyed in different yellow shades.

#### EXPERIMENTAL

Preparation of tetra-amido-diphenyl:—This compound was prepared according to the method of Strakosch<sup>16</sup>

(1) 3-4:3'-4' diphenyl-di  $\beta$ -naphthoquinoxaline:—A solution of 8.4 gram of  $\beta$ -naphthoquinone was mixed with a solution of 1 gram of tetraamidodiphenyl and refluxed for three hours. The precipitate obtained (yield 1.1 gram) was dried and recrystallised from acetone. It is soluble in acetone and sulphuric acid. ( $C_{22}H_{18}N_4$  requires N=12.23%, found N=12.35%),

(2) 3-4:3'-4' diphenyl-di-acenaphthoquinoxaline:—A solution of one gram. of acenaphthoquinone was mixed with a solution of one gram of tetra-amido-di-phenyl; refluxed for four and a half hours and the compound obtained (1.3 gr.) was recrystallised from a large volume of boiling alcohol. It is slightly soluble in pyridine and chloroform and freely soluble in conc. sulphuric acid. ( $C_{36}H_{18}N_4$  requires  $N=11.07\%$ , found  $N=10.72\%$ ).

(3) 3-4:3'-4' di-phenyl-di-phenanthraquinoxaline:—A solution of .7 gram of phenanthraquinone was mixed with a solution of one gram of tetra-amido-diphenyl and the reaction mixture was refluxed for three hours. The compound obtained (a gram) dissolves in boiling pyridine and conc. sulphuric acid. ( $C_{40}H_{14}N_4$  requires  $N=10.40\%$ , found  $N=9.1\%$ ).

(4) 3-4:3'-4' diphenyl-di-isatin-quinoxaline:—A red precipitate was obtained on mixing a solution of .9 gram of isatin and another solution of one gram of tetra-amidodiphenyl. The mixture was refluxed for three hours and the precipitate separated and dried (yield 1.5 grms.). It is soluble in pyridine and acetone and conc. sulphuric acid. ( $C_{28}H_{16}N_6$  requires  $N=18.91\%$ , found  $N=19.35\%$ ).

(5) 3-4:3'-4' di-phenyl-di-carboxyl-di-benzylquinoxaline: A mixture of a solution of 1.5 gram. of  $\alpha$ - $\alpha'$  dicyanodiketodibenzyl and another solution of 1 gram of tetra-amidodiphenyl was refluxed for eight hours and then allowed to stand over two days after a little water having been added to produce turbidity. A fine crystalline precipitate was thus obtained which dissolves in glacial acetic acid and recrystallises on dilution (yield .9 gram.). This yellowish brown compound melts at  $235^{\circ}$ - $236^{\circ}$  C. and is freely soluble in boiling alcohol, acetone, pyridine, caustic soda and sulphuric acid. ( $C_{18}H_{14}N_4O_8$  requires  $N=7.05\%$ , found  $N=8.44\%$ ).

(6) 3-4:3'-4' diphenyl-di-meta-nitroisatinquinoxaline:—A solution of one gram of meta-nitroisatin and another solution of .9 gram of tetra-amido-diphenyl was refluxed for five hours and the precipitate obtained was separated and dried (yield 1.35 grams.). The brownish red compound is soluble in hot acetone, pyridine, caustic soda and sulphuric acid.

( $C_{28}H_{14}N_8O_4$  requires  $N=21.29\%$ , found  $N=20.45\%$ ).

(7) 3-4:3'-4' diphenyl-di-parabanquinoxaline:—A mixture of the solutions of 1.4 grams of parabanic acid and 1.9 grams of tetra-amidodiphenyl was refluxed for four hours. The precipitate began to make its appearance towards the end of the reaction. The solution was allowed to stand

overnight and the compound separated (yield .6 gram.). The substance was recrystallised from boiling alcohol and it is soluble in pyridine, caustic soda, and sulphuric acid ( $C_{18}H_{10}N_8O_4$  requires  $N=30.27\%$ , found  $N=29.91\%$ ).

(8) 3-4:3'-4' diphenyl-dialloxanquinoxaline:—A reaction mixture of a solution of .6 gram of alloxan in a comparatively large volume of water and another solution of one gram of tetra-amidodiphenyl was refluxed for eight hours after adding some sodium acetate to it. The precipitate separated on cooling (yield .5 gram). It was recrystallised from alcohol and it is soluble in acetone, pyridine, caustic soda and sulphuric acid. ( $C_{20}H_{10}N_8O_4$  requires  $N=26.29\%$ , found  $N=25.93\%$ .)

(9) 3-4:3'-4' diphenyl-tetramethylquinoxaline:—.48 gram of diacetyl was dissolved in alcohol and to this was added a solution of one gram of tetra-amidodiphenyl and the mixture was refluxed for four hours. The precipitate which separates towards the end of the reaction was collected and dried. (yield .65 gram.) This jet black compound was recrystallised from glacial acetic acid. It is soluble in concentrated sulphuric acid. ( $C_{20}H_{14}N_4$  requires  $N=17.84\%$ , found  $N=17.9\%$ .)

(10) Diquinoxaline or 3-4:3'-4' diphenylquinoxaline:—A solution of .4 gram of glyoxal was added to another solution of one gram of tetraamidodiphenyl and the mixture was refluxed for five hours. The solution was filtered hot and water was gradually added with vigorous stirring to produce turbidity. The jet black crystals (yield .5 gram) were recrystallised from glacial acetic acid. They are slightly soluble in pyridine and dissolve in concentrated sulphuric acid. ( $C_{16}H_{10}N_4$  requires  $N=21.7\%$ , found  $N=20.92\%$ ).

(11) 3-4:3'-4' diphenyl-di-orthonitrophenyl-di-hydro-imidazole:—A mixture of a solution of 2 grams of orthonitrobenzaldehyde and another solution of 2 grams of tetra-amidodiphenyl was refluxed for eight hours and the contents were poured in cold water. The precipitate which on drying turned somewhat resinous was repeatedly extracted with boiling alcohol and the compound was obtained by the concentration of this alcoholic solution. The dark brown substance obtained melts at  $221^{\circ}$ – $222^{\circ}$  C. It is soluble in acetone, pyridine and sulphuric acid. ( $C_{26}H_{14}N_{10}O_4$  requires  $N=17.57\%$ , found  $N=17.35\%$ .)

(12) 3-4:3'-4' diphenyl-diparatolyl-dihydroimidazole:—A solution of .5 gram of paratolylaldehyde was mixed with a solution 1.5 grams of tetra-amido-diphenyl and the mixture was refluxed for seven hours. The contents

were poured in water, the precipitate separated and purified by crystallisation from alcohol (yield 1 gram) It is soluble in pyridine, acetone and sulphuric acid. ( $C_{25}H_{26}N_4$  requires  $N = 13.4\%$ , found  $N = 12.91\%$ .)

(13) 3-4:3'-4' diphenyl-diorthochlorophenyl-dihydroimidazole:—A solution of 5 gram of orthochlorobenzaldehyde was added to a solution of 1.5 grams of tetra-amidodiphenyl, the mixture refluxed for six hours and the contents were poured in cold water. The compound was recrystallised from alcohol. The brown crystalline powder is soluble in pyridine, acetone and sulphuric acid. ( $C_{26}H_{20}N_4Cl_2$  requires  $Cl = 15.47\%$ , found  $Cl = 15.4\%$ .)

(14) 3-4:3'-4' diphenyl dihydro-phenazine or bisdihydrophenazine:—A solution of 1.2 grams of catechol was treated with a solution of 2 grams of tetra-amidodiphenyl and the reaction mixture was refluxed for five hours. On dilution, it gave a precipitate which was dried and recrystallised from glacial acetic acid. It is soluble in pyridine and conc. sulphuric acid. ( $C_{24}H_{18}N_4$  requires  $N = 15.47\%$ , found  $N = 15.12\%$ .)

N. N. In all cases where the solvent is not mentioned, the reaction was carried in glacial acetic acid medium.

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